PB81-236135

Proceedings of the Joint Symposium on Stationary Combustion NOx Control. Volume III: Utility Boiler NOx Control by Flue Gas Treatment

(U.S.) Industrial Environmental Research Lab.
Research Triangle Park, NC

Prepared for

Eletric Power Research Inst.
Palo Alto, CA

Aug 81

U.S. Department of Commerce
National Technical Information Service
NTIS

PB81-236135

IERL-RTP-1085a

Proceedings of the Joint Symposium on Stationary Combustion NO_x Control

Volume III

Utility Boiler NO_x Control

by Flue Gas Treatment

Symposium Cochairmen
Robert E. Hall, EPA
and
J. Edward Cichanowicz, EPRI

Program Element No. N130

Prepared for

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Research and Development
Washington, D.C. 20460
and
ELECTRIC POWER RESEARCH INSTITUTE
3412 Hillview Avenue
Palo Alto, California 94303

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INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA 22161

i

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. REPORT NO.
EPA-600/9/81/028c

2.
Proceedings

3. RECIPIENT'S ACCESSION NO.
PB81 236135

4. TITLE AND SUBTITLE
Proceedings of the Joint Symposium on Stationary Combustion NO_x Control. Vol. III Utility Boiler NO_x Control by Flue Gas Treatment

5. REPORT DATE
August 1981

6. PERFORMING ORGANIZATION CODE

7. AUTHOR(S)
Symposium Cochairmen: Robert E. Hall (EPA) and J.E. Cichanowicz (EPRI)

8. PERFORMING ORGANIZATION REPORT NO.
IERL-RTP-1085a

9. PERFORMING ORGANIZATION NAME AND ADDRESS
See Block 12.

10. PROGRAM ELEMENT NO.
EHE624

11. CONTRACT/GRANT NO.
NA (Inhouse)

12. SPONSORING AGENCY NAME AND ADDRESS
EPA, Office of Research and Development
Industrial Environmental Research Laboratory
Research Triangle Park, NC 27711

13. TYPE OF REPORT AND PERIOD COVERED
Proceedings; 10/6-9/80

14. SPONSORING AGENCY CODE
EPA/600/13

15. SUPPLEMENTARY NOTES EPA-600/7-79-050a through -050e describe the previous symposium.

16. ABSTRACT

The proceedings document the approximately 50 presentations made during the symposium, October 6-9, 1980, in Denver, CO. The symposium was sponsored by the Combustion Research Branch of EPA's Industrial Environmental Research Laboratory, Research Triangle Park, NC, and the Electric Power Research Institute (EPRI), Palo Alto, CA. Main topics included utility boiler field tests; NOx flue gas treatment; advanced combustion processes; environmental assessments; industrial, commercial, and residential combustion sources; and fundamental combustion research. This volume relates to the treatment of flue gases from utility boilers to control NOx emissions.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group	
Pollution	Flue Gases	Pollution Control	13B	
Combustion	Engines	Stationary Sources	21B	21K
Nitrogen Oxides		Environmental Assessment	07B	
Boilers			13A	
Tests			14B	
Assessments				

18. DISTRIBUTION STATEMENT Release to Public	19. SECURITY CLASS *(This Report)* Unclassified	21. NO. OF PAGES 276
	20. SECURITY CLASS *(This page)* Unclassified	22. PRICE

EPA Form 2220-1 (9-73)

([illegible] COVER 4)

PREFACE

These proceedings document more than 50 presentations given at the Joint Symposium on Stationary Combustion NO_x Control held October 6-9, 1980 at the Stouffer's Denver Inn in Denver, Colorado. The symposium was sponsored by the Combustion Research Branch of the Environmental Protection Agency's (EPA) Industrial Environmental Research Laboratory-Research Triangle Park and the Electric Power Research Institute (EPRI). The presentations emphasized recent developments in NO_x control technology. Cochairmen of the symposium were Robert E. Hall, EPA, and J. Edward Cichanowicz, EPRI. Introductory remarks were made by Kurt E. Yeager, Director, Coal Combustion Systems Division, EPRI, and the welcoming address was given by Roger L. Williams, Regional Administrator, EPA Region VIII. Stephen J. Gage, Assistant Administrator for Research and Development, EPA, was the keynote speaker. The symposium had 11 sessions:

I:	NO_x Emissions Issues Michael J. Miller, EPRI, Session Chairman
II:	Manufacturers Update of Commercially Available Combustion Technology Joshua S. Bowen, EPA, Session Chairman
III:	NO_x Emissions Characterization of Full Scale Utility Powerplants David G. Lachapelle, EPA, Session Chairman
IV:	Low NO_x Combustion Development Michael W. McElroy, EPRI, Session Chairman
Va:	Postcombustion NO_x Control George P. Green, Public Service Company of Colorado, Session Chairman
Vb:	Fundamental Combustion Research Tom W. Lester, EPA, Session Chairman
VI:	Status of Flue Gas Treatment for Coal-Fired Boilers Dan V. Giovanni, EPRI, Session Chairman
VII:	Small Industrial, Commercial, and Residential Systems Robert E. Hall, EPA, Session Chairman
VIII:	Large Industrial Boilers J. David Mobley, EPA, Session Chairman
IX:	Environmental Assessment Robert P. Hangebrauck, EPA, Session Chairman
X:	Stationary Engines and Industrial Process Combustion Systems John H. Wasser, EPA, Session Chairman
XI:	Advanced Processes G. Blair Martin, EPA, Session Chairman

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EMPIRICAL EVALUATION OF SELECTIVE CATALYTIC REDUCTION AS AN NO_x CONTROL TECHNIQUE

By:

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ABSTRACT

Selective catalytic reduction (SCR) has been proposed as a technique for control of NO_x emissions to levels significantly below those mandated by NSPS for coal-fired utility steam generators. EPRI is conducting an empirical assessment of the feasibility and cost-effectiveness of SCR, using a pilot scale system at the EPRI Arapahoe Emission Test Facility to simulate authentic coal-fired utility operating conditions. The program is a logical extension to earlier EPRI work defining economic feasibility of postcombustion control, and complementary to other pilot scale studies in the United States and Japan.

The test program was initiated in September 1980 on a facility capable of treating 5,000 scfm of coal-fired flue gas, equivalent to 2.5 MW of electrical generating capacity. The facility employs a regenerative air heater in series with a catalytic reactor to assess potential impacts on air heater performance. The tests will focus on four major issues important to the evaluation of SCR technology: (1) process performance as defined by NO_x removal capabilities at conditions representative of authentic utility application; (2) process operating demands including the need for monitoring and control systems, consumables such as ammonia, energy (pressure drop and auxiliary power), operating and maintenance requirements to maintain process performance, catalyst lifetime; (3) environmental impacts due to emissions of residual ammonia, SO_3, and sulfates and bisulfates of ammonia; and the potential effects on SO_2 and particulate control; and (4) systemwide operating effects such as increased operating and maintenance of downstream surfaces (particularly the air heater), heat rate penalty, limitations in load-following, etc.

Results are presented for the initial tasks dealing with the evaluation of measurement techniques, and preliminary data describing reactor and air heater performance.

ACKNOWLEDGEMENTS

The authors wish to acknowledge the efforts of the many individuals involved at all levels in this project. Dave Naulty and Mike Moora of Stearns-Roger, Inc., were primary individuals responsible for the procurement and construction of the pilot facility. Assistance from Kawasaki Heavy Industries was provided by Senji Niwa. Operation and maintenance of the pilot plant was managed by Jim Parsons and John Serdinsky of Kaiser Engineers. Testing was conducted by Gary Shiomoto, Larry Muzio, and Robert Pease of KVB. Finally, the key role played by EPRI staff members at the Arapahoe Emission Test Facility, namely Richard Hooper and Lou Rettenmaier, is most gratefully appreciated.

EMPIRICAL EVALUATION OF SELECTIVE CATALYTIC REDUCTION AS AN NO_x CONTROL TECHNIQUE

INTRODUCTION

The Environmental Protection Agency (EPA) has cited as goals the reduction of New Source Performance Standards (NSPS) for oxides of nitrogen (NO_x) emissions from coal-fired utility power plants from current levels of 0.5-0.6 lbs/million Btu's (depending on coal properties) to 0.2. Under certain circumstances, coal-fired power plants could be required to further reduce NO_x emissions below NSPS levels, such as in Air Quality Regions (AQRs) in noncompliance with the proposed Federal ambient NO_2 standard, or to prevent significant deterioration of Class I AQRs. Such regulations have already been promulgated for certain classes of oil- and gas-fired steam generators in California's South Coast Air Basin. Postcombustion methods of NO_x control (alternatively known as flue gas treatment) have been proposed to achieve such stringent standards for both clean fuels (such as natural gas and fuel oil) as well as coal. Accordingly, the electric utility industry through EPRI is conducting an empirical evaluation of the feasibility of postcombustion NO_x control processes for coal-fired utility boilers.

Development efforts in postcombustion control have been conducted predominantly in Japan and in the last decade. This is due to stringent Japanese emissions standards; and to the predominant use in that country of natural gas and fuel oil, which due to low ash and sulfur content, offer greater prospects for successful application. Extensive laboratory and pilot scale work has been conducted for these "clean" fuels, and recently has been expanded to consider coal. Most of this development work is typified by well-controlled operating conditions necessary for fundamental process development, but not representative of authentic utility application. Extrapolation of Japanese pilot plant experience with coal to U.S. utility systems is complicated not only by significant scale factors but also by differences in coal properties (i.e., ash characteristics and chemistry) between coals used in Japan and the United States. Experience with full-scale utility systems is limited exclusively to clean fuels, and thus does not reflect the problems inherent to high particulate loading, SO_2 concentration, and ash chemistry.

SUMMARY OF RELEVANT EPRI EXPERIENCE

Prior work conducted by EPRI has consisted of preliminary technical and economic assessments of the feasibility of postcombustion processes for U.S. utility coal-fired application. In two initial studies (1,2), approximately 50 processes were categorized as to technical basis and screened according to potential merits and economics of operation. Follow-on studies considered a limited number of processes in more detail through design studies of specific applications. Seven postcombustion processes were evaluated by the Tennessee Valley Authority (TVA) through a jointly funded program with the EPA; the results identified the dry, ammonia-based selective catalytic reduction (SCR) process as a leading candidate for near-term commercial acceptance (3,4). A further study conducted by Stearns-Roger for EPRI examined two dry SCR processes in addition to the Shell/UOP simultaneous SO_2/NO_x and the Exxon Thermal $DeNO_x$ process (5). Both the TVA and Stearns-Roger studies identified for each process (a) the technical merits and potential obstacles to application to new coal-fired utility boilers, and (b) the economics of operation, necessarily limited to the costs of process equipment procurement, installation, and operation. Only the most obvious impacts of SCR on powerplant design and operation that could be identified with the limited level of experience were included. A study similar in scope and depth has been recently completed, also by Stearns-Roger, to assess costs of retrofit application of SCR systems from two process vendors to an 80 MW boiler (6).

One of these preliminary investigations (5) has identified the minimum cost of SCR application to new 500 MW coal-fired power plants, for the case of 90% NO_x removal from initial NO_x concentrations representative of present NSPS levels. These results, summarized in Table 1 for the Kawasaki Heavy Industries (KHI) process, are preliminary and therefore incomplete as all potential impacts on plant operation attributable to SCR have not been identified. This can only be accomplished through a pilot-scale evaluation of a commercially representative system on a coal-fired utility boiler at operating conditions that reflect authentic utility application. The remainder of this paper provides a status report of such an effort at the EPRI Arapahoe Emission Test Facility.

POTENTIAL SCR EFFECTS ON FLUE GAS FROM COAL

Figure 1 presents a diagrammatic path for the potential fates of the major nitrogen and sulfur species in flue gas subjected to SCR treatment. The temperature range of interest spans from typical catalyst temperatures (specific for one process, but representative of other SCR processes) to typical air heater exit temperatures. The process inputs are indicated at the reactor inlet as are the desired products of the NH_3/NO_x reduction reactions, and the residual and by-product emissions of NH_3 and SO_3. As the flue gas proceeds through the reactor and downstream equipment and cools, the following fates of major nitrogen and sulfur species are possible:

(1) SO_2 can be

(a) oxidized to SO_3 within the reactor (usually 1-5% inlet SO_2);

(b) collected in the SO_2 scrubber and converted to the liquid or solid scrubber effluent (ultimately to be transferred to the scrubber waste disposal site);

(c) emitted

(2) SO_3 can

(a) be collected in the scrubber and transferred to the liquid or solid scrubber effluent, and ultimately to scrubber waste disposal site;

(b) be emitted, either in gas phase or liquid if the dew point is reached;

(c) condense or be adsorbed on internal surfaces; or on fly ash, and thus collect in the particulate control device (and ultimately the ash disposal site), or penetrate the control device.

(3) NH_3 can remain in gaseous form and be

(a) emitted;

(b) absorbed in the scrubber (disposal site);

(c) absorbed by fly ash and thus collect in the particulate control device (ash disposal site), or penetrate the control device.

(4) NH_3 and SO_3 can combine and form (depending on relative NH_3, SO_3, and H_2O concentration, and temperature) ammonium sulfate or ammonium bisulfate (liquid or solid phase) which can:

(a) pass through the air heater and collect in either the scrubber or particulate control device (disposal site), or penetrate the control device;

(b) impact and lodge on air heater or other internal surfaces and subsequently be removed by sootblowing and endure the fate described in (a), or be removed through water washing and transferred to the wash effluent;

(c) be absorbed by fly ash, and collect in the particulate control device (ash disposal site), or penetrate the control device and be emitted.

Based upon these potential paths, at least four ultimate fates for nitrogen and sulfur residual and by-product species can be identified:

- airborne emission
- retained on surfaces within system (possibly removed through routine washing maintenance procedures)
- SO_2 scrubber/effluent disposal site

- particulate collection device/ash disposal site

Each of these affected areas has been the focus of considerable attention, and the added impact of SCR by-products could raise new issues requiring additional corrective action.

Table 2 presents a compilation of the issues of concern attributable to SCR that must be addressed before the feasibility of SCR can be assessed. These issues are attributable to the operation and maintenance of both the SCR process and the entire power plant, as determined by the four potential fates of major nitrogen and sulfur species. These issues are categorized as to (a) process performance, (b) process operating demands, (c) environmental impacts, and (d) system-wide operating effects. For these issues of concern, the data necessary from the test program for an empirical assessment is cited, as well as the economic implications for power plant operation.

This table has been used as the basis for the test plan which is presented in a later section.

DESCRIPTION OF FACILITY

The empirical assessment of SCR for coal-fired utility boilers is being conducted at the EPRI Arapahoe Emission Test Facility. This facility, operated in cooperation with the Public Service Company of Colorado (PSCCo), is located in Denver, and was commissioned in 1977 to support the testing and evaluation of advanced particulate control technology. The direction of work at Arapahoe has been expanded to include the present evaluation of NO_x control, and in the future (1981) will involve testing and evaluation of SO_2 scrubbers, and address the integrated design of all components in an emission control train.

In December of 1978, EPRI contracted with KHI to provide a 2.5 MW (electrical equivalent) SCR pilot plant. The selection of KHI does not represent an endorsement or preference by EPRI for that process, but rather an acknowledgment that the process is representative of SCR systems currently available. Similarly, the C-E Air Preheater Company was selected to supply for the same flow capacity a Ljungstrom regenerative air heater which is considered representative of air heater designs in current and anticipated usage.

Figure 2 presents an outline arrangement of the pilot scale reactor and air heater. Coal-fired flue gas is extracted from PSCCo Unit No. 4 at the economizer exit (approximately 650°F) and ducted to a control valve, venturi for flow measurement, and an electrical resistance heater for control of inlet gas temperature (not shown in figure). The ammonia vaporizer and injection assembly is located at the base of the flue duct riser. The catalyst is housed in three reactor modules arranged in series, each with an equivalent quantity of catalyst. The regenerative air heater is located downstream, and oriented for a horizontal axis of rotation.

Specifications for the KHI SCR process are presented in Table 3. The major active catalytic ingredient is vanadium pentoxide, stabilized or supported in a titanium dioxide base. The material is formed in cylindrical shapes, and arranged as shown in Figure 3, with spacing selected to minimize particulate accumulation and plugging. Total catalyst volume is 120 ft^3, corresponding to a design space velocity of 2500 hr^{-1} at the rated flow capacity of 5000 scfm.

The ammonia vaporizer and delivery system have been designed to simulate the hardware and operation of the full-scale systems proposed by KHI for commercial application. The ammonia is transported from a storage tank to the vaporizer, and then mixed with carrier air (approximately 20/1 air/NH_3 ratio) preheated to 120°F and injected into the flue gas via a mixing grid.

Design specifications are presented in Table 4 for the regenerative air heater, which is designed to simulate the heat transfer and sootblowing characteristics of full-scale utility operation. In this context, the temperature history of the wheel elements and the mass flux per unit time of sootblowing media have been selected to represent full-scale values. An evaluation of the tolerance and susceptibility of three materials of wheel basket construction to plugging/fouling/corrosion by fly ash and SCR residual and by-product emissions is possible. Stainless steel, hard-rolled steel with baked enamel finish, and Corten are all employed in several arrangements of wheel basket construction.

The facility is equipped with process control systems to (a) facilitate pilot plant operation and provide for well-controlled test conditions, and

(b) evaluate typical control systems that could be necessary for commercial acceptance of SCR. Control systems for the catalytic reactor are necessary to adjust the mass NH_3/NO_x ratio according to changing input reactor conditions.

Five control modes will be employed at Arapahoe to both facilitate pilot plant operation and simulate the actual conditions of SCR operation in a full-scale commercial application. Each of these modes is listed in Table 5, which identifies for each the continuous on-line processing required, and EPRI experience to date. The least on-line processing is required for the first two modes that maintain either a specified NH_3/NO_x ratio or NO_x removal efficiency. The most processing is required for the latter three (which all require continuous measurement of NH_3 at the reactor outlet) to limit the mass of NH_3 injection to an amount that maintains NH_3 carryover below a prescribed level.

Continuous measurement of NH_3 in flue gas is being attempted with a special-purpose sampling probe/catalytic converter. This apparatus selectively oxidizes NH_3 to NO, permitting NH_3 concentration to be inferred from the difference in NO measurement before and after NH_3 oxidation. A schematic of the sampling probe and the analytical system is presented in Figure 4. Results to date with this system have not been successful, thus prohibiting any of the latter three control modes in Table 5 to be employed.

Two control systems are employed to drive air heater operation, and are primarily used to provide well-controlled test conditions over the long time periods required for assessment of air heater operation. Both controls vary air side flow rate and static pressure to maintain a prescribed (a) flue gas exit temperature, and (b) air side/gas side pressure differential (i.e., leakage) independent of flue gas inlet temperature, pressure, and flow rate.

TEST PLAN

A test plan has been developed to fulfill the project objectives in a time- and cost-effective manner. Table 6 defines the tasks of the test plan and identifies for each the primary and secondary variables, and those parameters that remain constant. As indicated in Table 6 the tests will evaluate the issues over the following operating conditions:

- baseline conditions, or those at or near the design conditions that could be encountered during commercial operation (Task 3);
- transient, or nonsteady conditions, simulating load swings, excess air changes, changes in boiler operating conditions, etc., (Task 4);
- upset conditions, characterized by high carbon monoxide (CO) and unburned hydrocarbon (HC) concentration, such as experienced in rapid shutdown or when encountering problems in fuel preparation (pulverizer or mill malfunctions) (Task 5);
- high particulate and SO_2 conditions, providing an approximation of the effects of ash loading and SO_2 concentration (Tasks 5, 6).

The test plan will also provide information regarding:

- catalyst lifetime, by documenting changes in catalyst performance over a 12 month period, employing well-controlled, repetitive performance tests at comparable operating conditions (Task 7), and
- air heater performance, consisting of both continuous analysis of daily performance data as well as repetitive tests at well-controlled, comparable test conditions (Task 8).

Tasks have been scheduled in a parallel sequence so as to maximize the utilization of the facility. The pilot facility operates on a 24 hour basis, with most parametric testing conducted during daytime hours when boiler conditions are generally steady-state and flue gas characteristics relatively constant. During evening hours, boiler conditions are generally not steady providing realistic transient conditions which can be used for evaluation of ammonia injection control schemes.

RESULTS

The initial phases of this project which have been completed (Tasks 1 and 2) are concerned with the procurement of the sampling/analytical equipment and development of appropriate test methodologies. Preliminary results at limited test conditions are available describing SCR reactor and air heater performance.

Tasks 1 and 2 efforts served to (a) verify conventional sampling/analytical techniques for major stable combustion products, and (b) evaluate a limited number of unconventional sampling/analytical techniques for the major species of concern (NH_3, SO_3, and $(NH_4)HSO_4$ and $(NH_4)_2SO_4$). Once the specific technique best suited for field testing was selected, the exact configuration of sampling trains and the specific sampling/analytical methodologies were determined. Detailed methodologies were identified for sampling gas phase NH_3 and total NH_4^+ (impinger collection with analysis by specific ion electrode) and gas phase SO_3 (sampling by controlled condensation, analysis by titration for $SO_4^=$ using lead perchlorate). These methodologies were determined for application at the reactor inlet, reactor outlet, and air heater outlet.

A summary of the preliminary results is presented in Figure 5, which indicates NO_x removal, NH_3 carryover, and SO_2 to SO_3 oxidation (as percent of inlet NO_x, NH_3, and SO_2, respectively). Results are displayed from KHI tests in Japan and preliminary Task 3 results from Arapahoe (data in parentheses). Results from Arapahoe are presented for one temperature; the temperature effect determined by KHI when available is indicated. Figure 5 indicates the results from KHI and preliminary Arapahoe results are similar.

Simultaneously with Tasks 2 and 3, a preliminary evaluation of the influence of SCR residual and by-product emissions on air heater performance was conducted. Initially, baseline performance was documented for over three weeks, while flue gas passed through the reactor but without ammonia injection. Measurements confirmed air heater performance (thermal efficiency and pressure drop) to be as predicted by C-E Air Preheater. Figure 6 presents the inlet/outlet gas side pressure differential as a function of continuous operating time for the baseline tests and after three weeks of operation with NH_3 and SO_3 in the flue gas. The significant increase in pressure drop (approximately 3-6 inches H_2O) was observed with sootblowing frequency maintained as recommended by the manufacturer, but with a reduced air pressure and mass flux (due to limitations in the air delivery system, since rectified). At the conclusion of the three week testing period with NH_3 injection, the pilot plant was removed from service and the wheel baskets disassembled (Figure 7), revealing significant accumulation of deposits at the cold-intermediate basket interface. Figure 8 shows a close-up of the deposit buildup on an intermediate basket face constructed of mild steel coated baked enamel. Qualitative analysis of a deposit sample indicated the presence of ammonia suggesting ammonium sulfate/bisulfate constituents.

FUTURE PLANS

The test plan is currently being carried out at the Arapahoe Emission Test Facility. It is anticipated that testing will be complete by Fall 1981.

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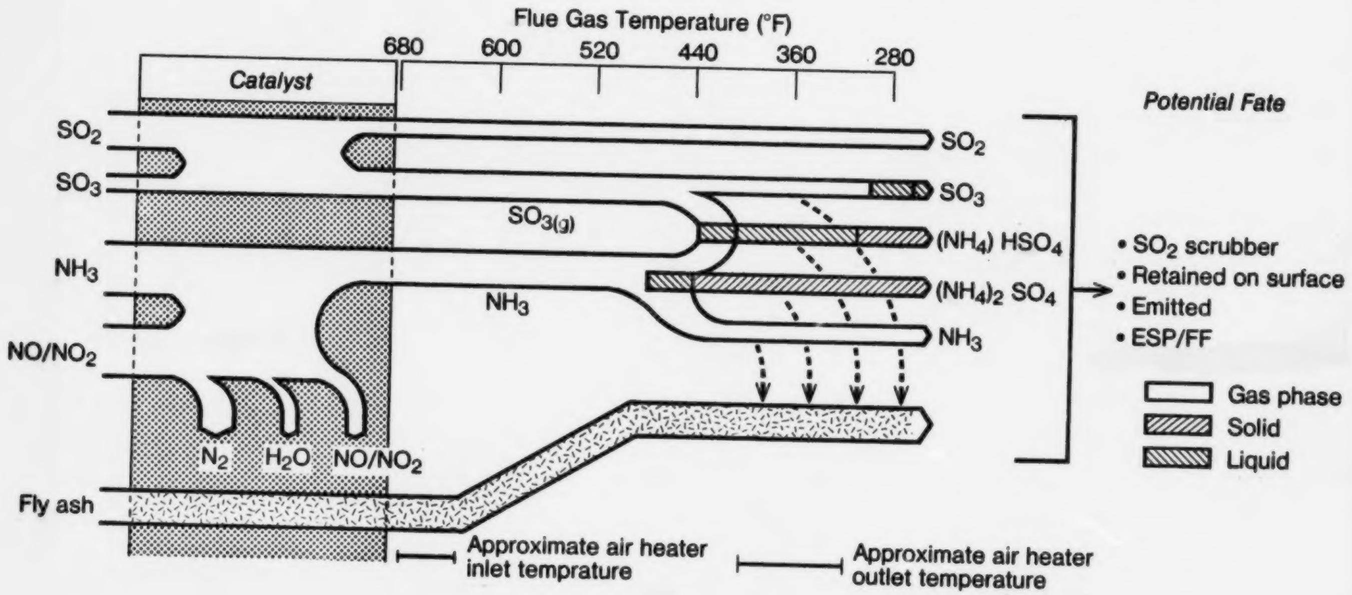
Flue Gas Temperature (°F)
680
600
520
440
360
280
Catalyst
SO2
SO3
NH3
NO/NO2
Fly ash
SO3(g)
NH3
N2
H2O
NO/NO2
SO2
SO3
(NH4) HSO4
(NH4)2 SO4
NH3
Potential Fate
• SO2 scrubber
• Retained on surface
• Emitted
• ESP/FF
Gas phase
Solid
Liquid
Approximate air heater inlet temprature
Approximate air heater outlet temperature

Figure 1. Fate of Major Nitrogen and Sulfur Species

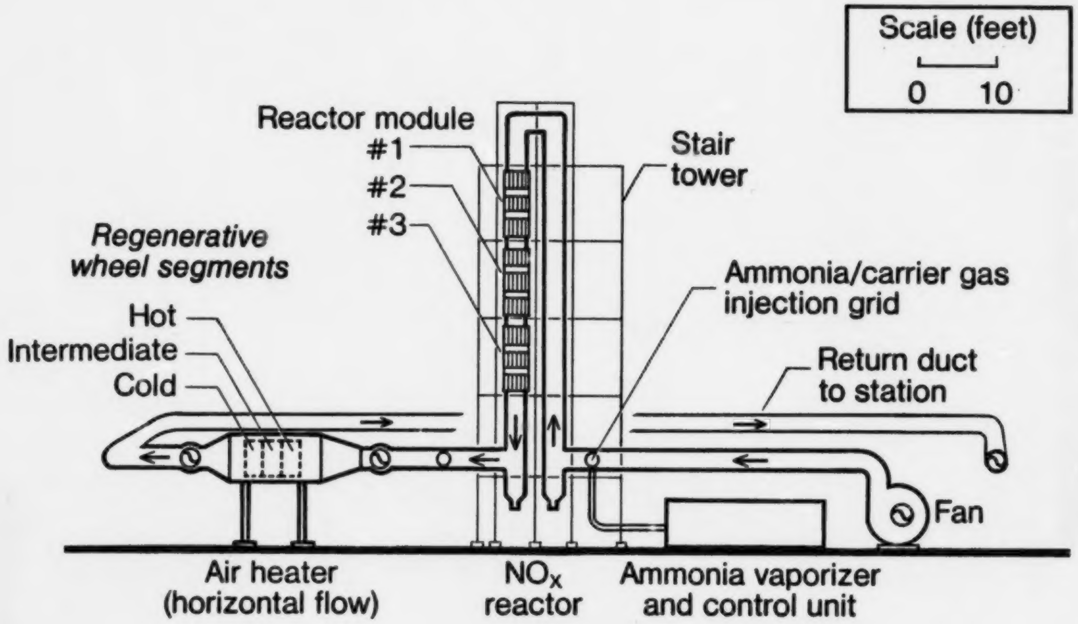
Scale (feet)
0 10
Reactor module
#1
#2
#3
Stair tower
Regenerative wheel segments
Hot
Intermediate
Cold
Ammonia/carrier gas injection grid
Return duct to station
Fan
Air heater (horizontal flow)
NO_x reactor
Ammonia vaporizer and control unit

Figure 2. NO_x Reactor and Air Heater

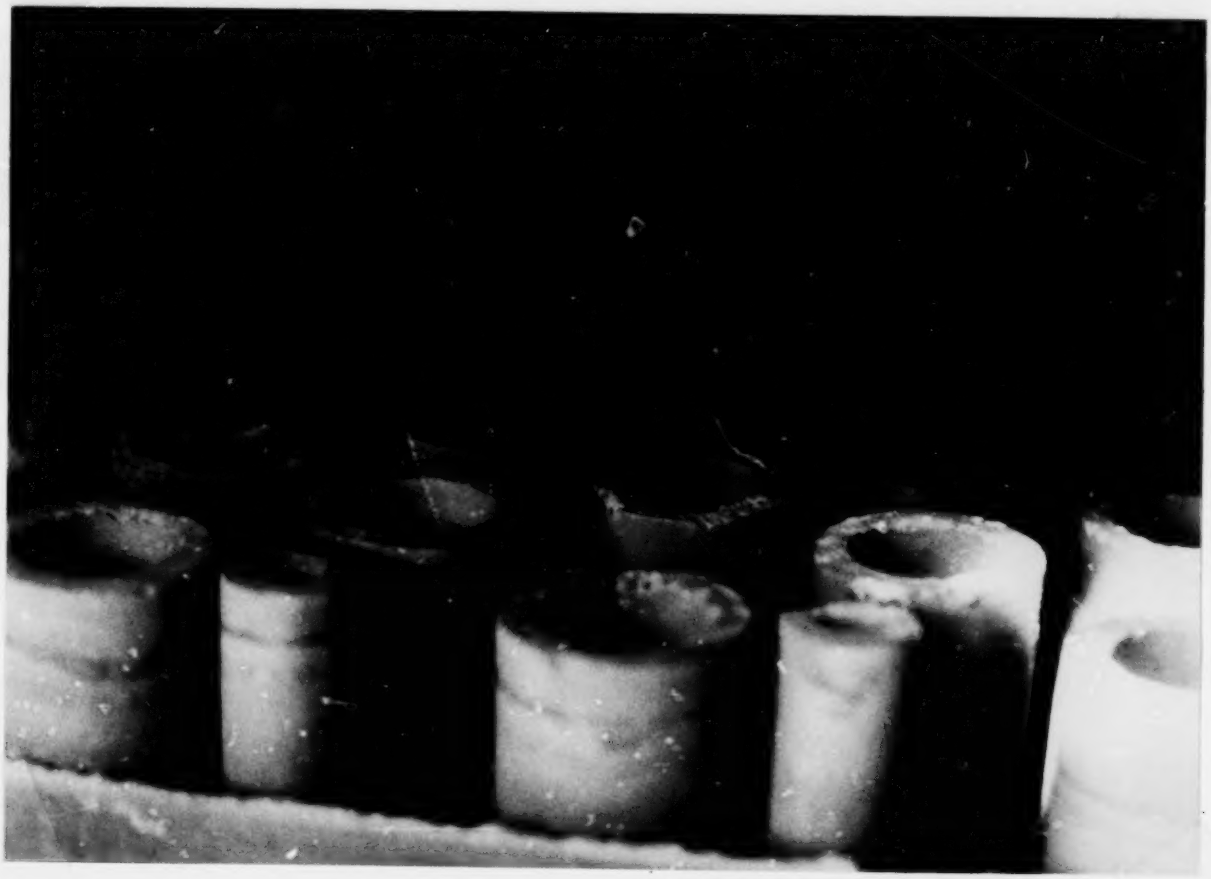

Figure 3. Cylindrical Shaped Catalyst

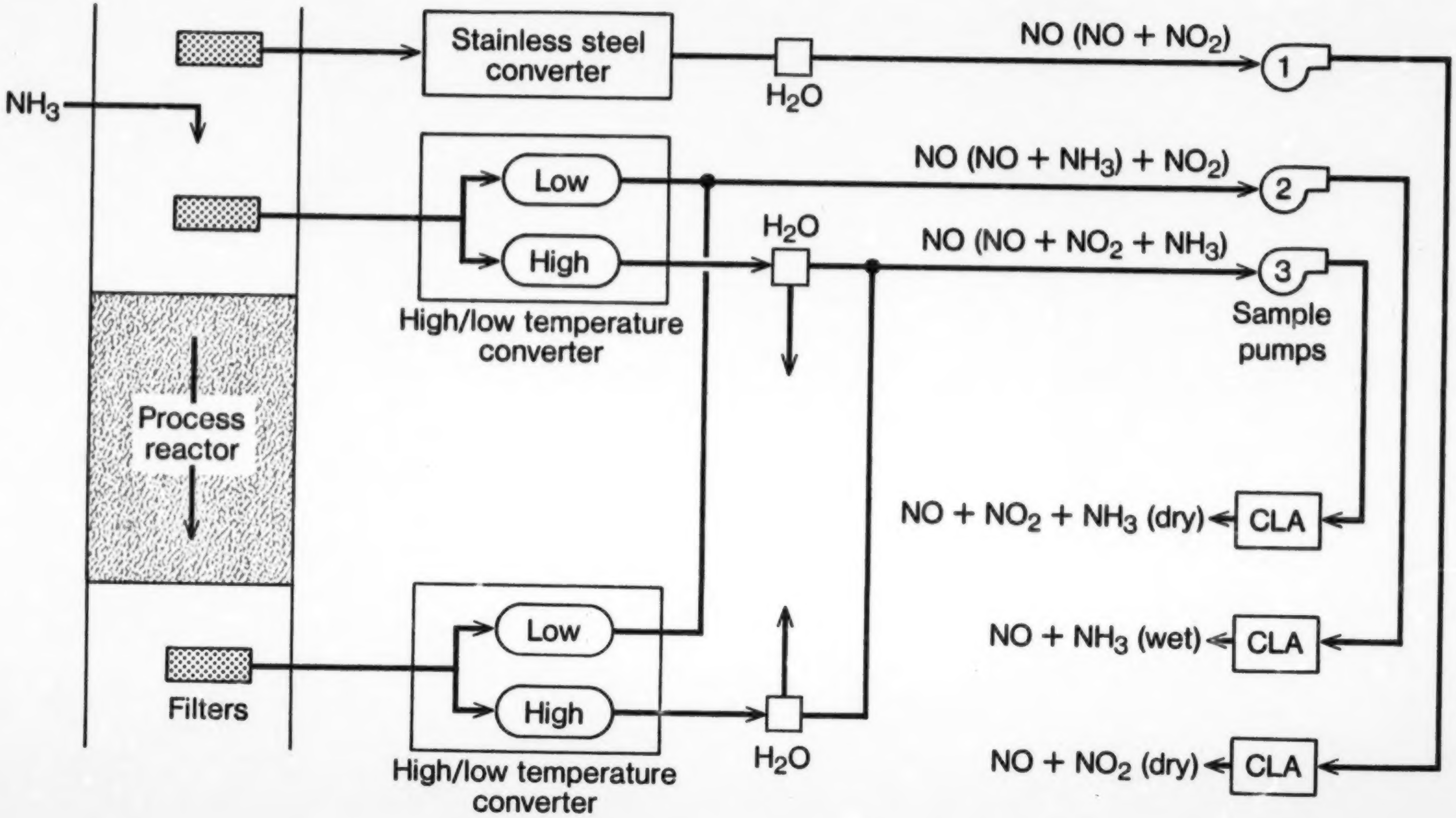
Stainless steel converter
NH$_3$
H_2O
NO (NO + NO$_2$)
1
Low
High
High/low temperature converter
H_2O
NO (NO + NH$_3$) + NO$_2$
2
NO (NO + NO$_2$ + NH$_3$)
3
Sample pumps
Process reactor
NO + NO$_2$ + NH$_3$ (dry)
CLA
Low
High
NO + NH$_3$ (wet)
CLA
Filters
High/low temperature converter
H_2O
NO + NO$_2$ (dry)
CLA

Figure 4. Continuous NH$_3$ Analyzer

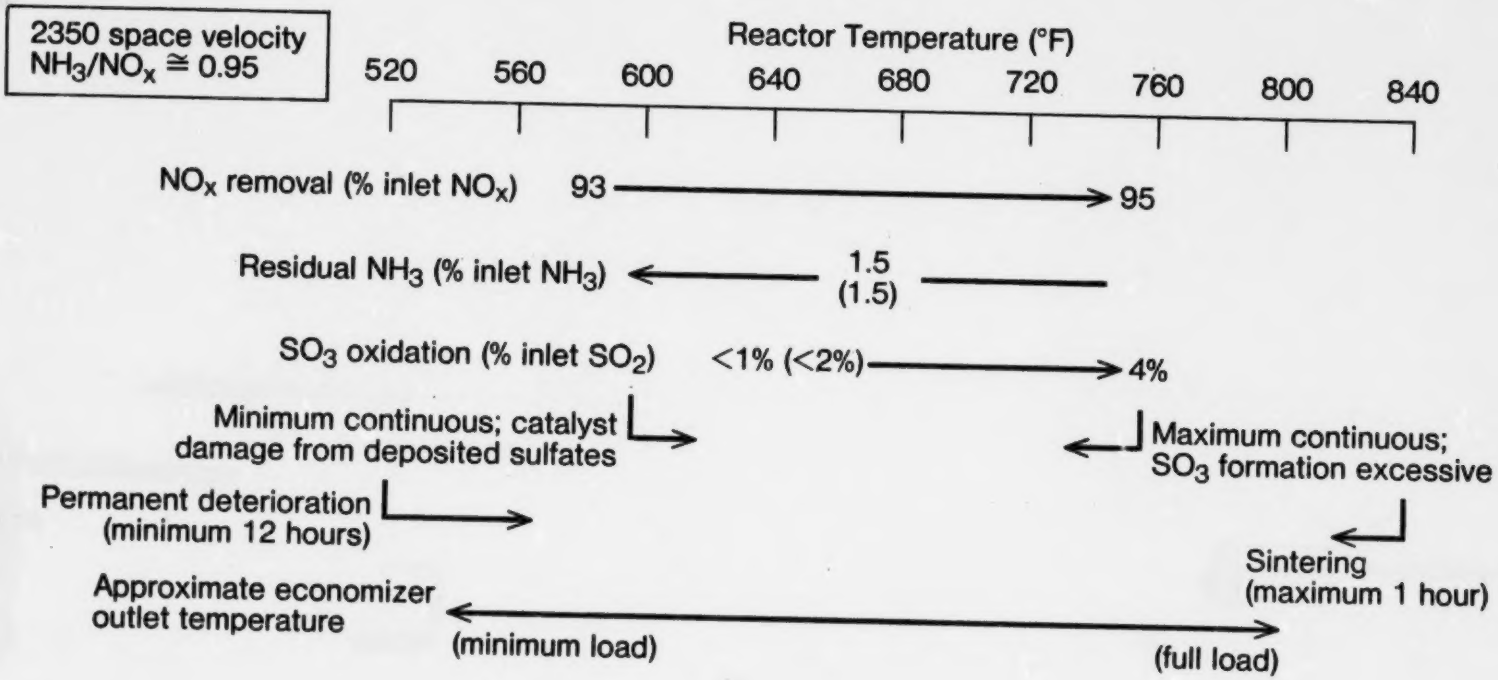
2350 space velocity
$NH_3/NO_x \cong 0.95$
Reactor Temperature (°F)
520
560
600
640
680
720
760
800
840
NO_x removal (% inlet NO_x)
93
95
Residual NH_3 (% inlet NH_3)
1.5
(1.5)
SO_3 oxidation (% inlet SO_2)
<1% (<2%)
4%
Minimum continuous; catalyst damage from deposited sulfates
Maximum continuous; SO_3 formation excessive
Permanent deterioration (minimum 12 hours)
Sintering (maximum 1 hour)
Approximate economizer outlet temperature
(minimum load)
(full load)

Figure 5. Influence of Reactor Temperature on Operation and Performance

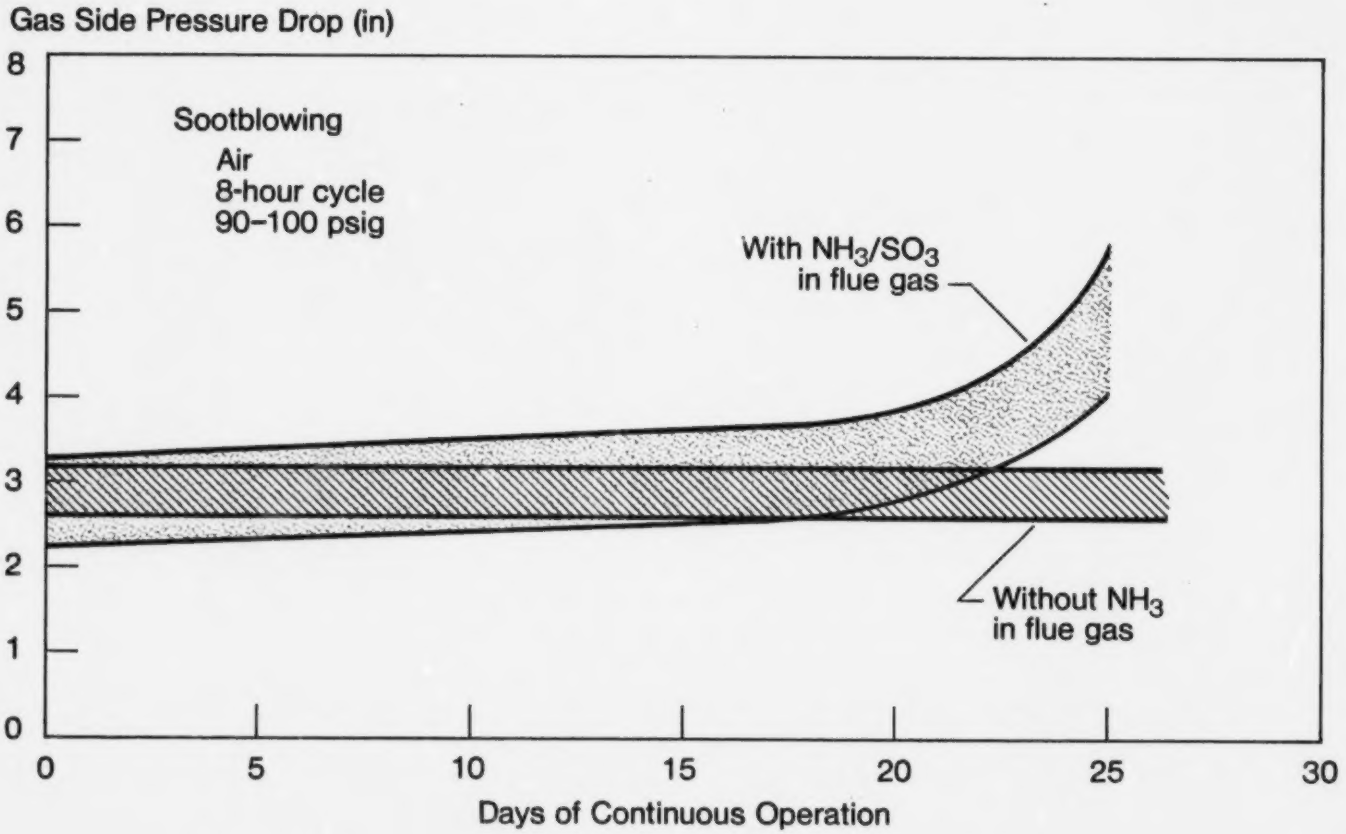

Figure 6. Influence of SO_3/NH_3 in Flue Gas on Air Heater Pressure Drop

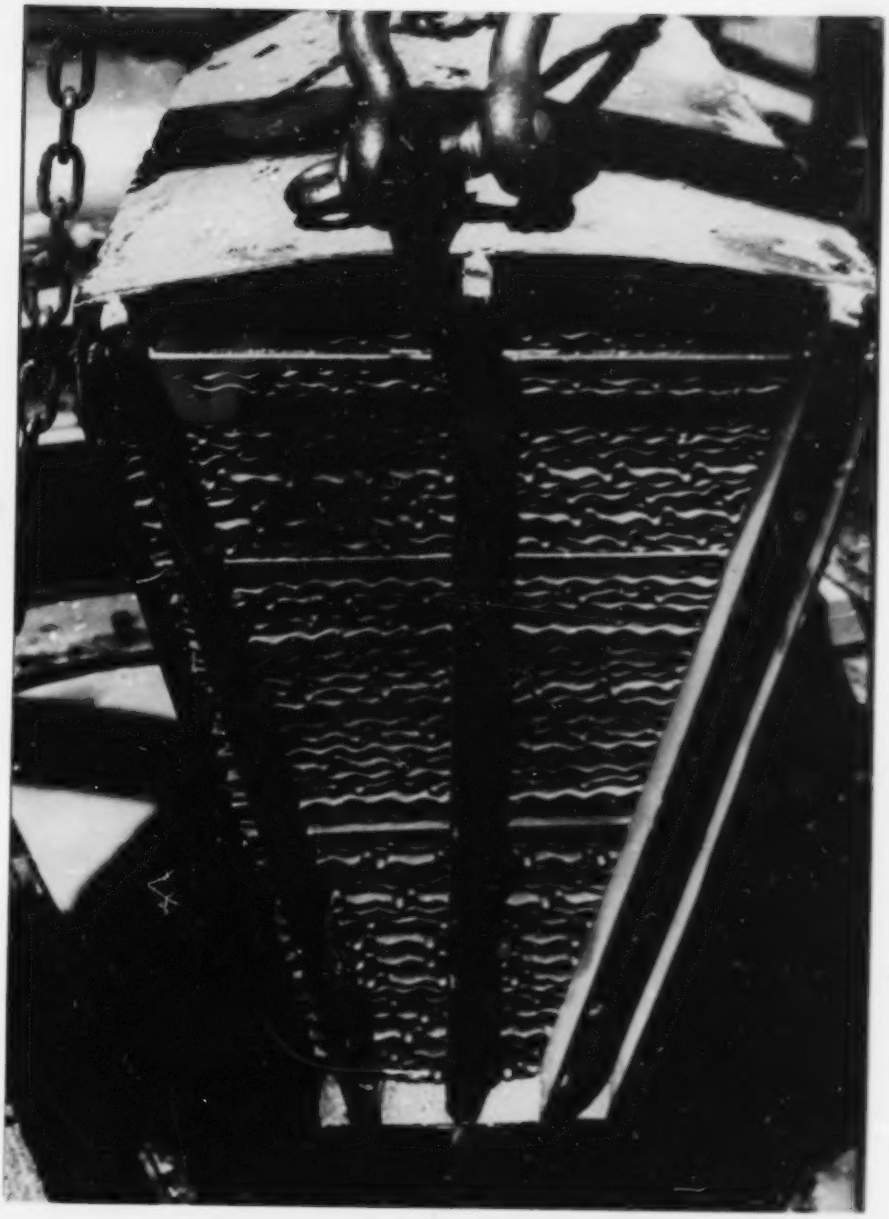

Figure 7. Regenerative Wheel Basket

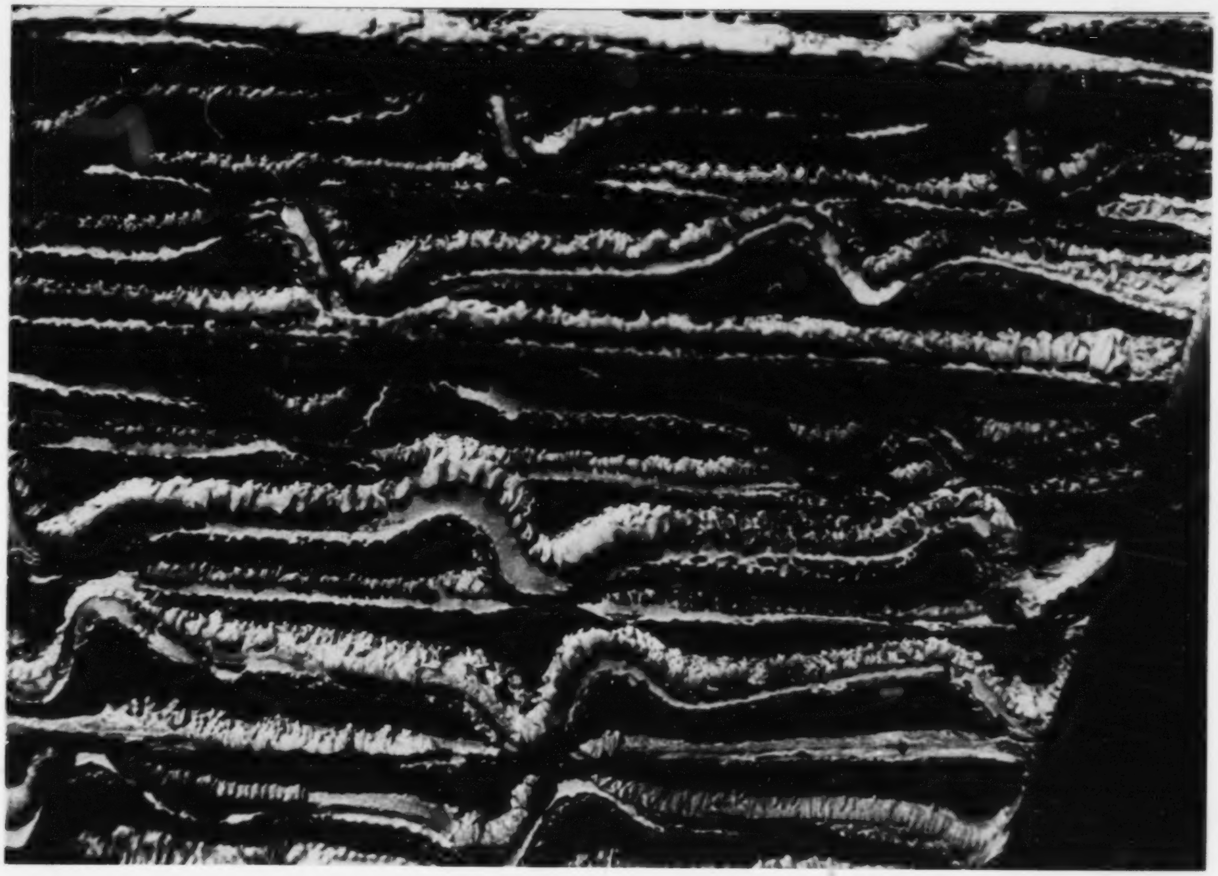

Figure 8. Deposit Accumulation on Regenerative Wheel Basket (cold face of intermediate segment)

TABLE 1

MINIMUM IDENTIFIABLE COSTS OF KAWASAKI HEAVY INDUSTRIES SELECTIVE CATALYTIC REDUCTION PROCESS

*CAPITAL REQUIREMENT:	41 - 60 $/Kw
*LEVELIZED OPERATING COST:	4.0 - 5.9 mills/kwh

Study Basis

Design Premises

- 90% NO_x removal (300 ppm @ 5% O_2 initial NO_x)
- 500 MW (net), 548 (gross) generating capacity
- Midwest location
- Economizer bypass employed for low loads (40 - 60% Maximum Rated Capacity)
- Low-sulfur, sub-bituminous coal (typical of Powder River Basin in Wyoming, Montana)

Economic Premises

- 1979 dollars
- Escalation factor 6% per annum
- Capital equipment inflation 6% per annum
- Levelizing period 30 years

* from Reference No. 5, "NO_x Control for Western Coal-Fired Boilers-Feasibility of Selected Postcombustion Systems"

Table 2

SCR OPERATING ISSUES

Operating Issues To be Addressed	Information Required	Economic Implications for Plant Operations
1. Performance (de-nitrification efficiency)	• inlet/outlet NOx	• quantity of catalyst required for given NOx emission level
2. Process Operating Demands		
a. ammonia	• ammonia demand vs SCR operating parameters	• cost of continuous ammonia supply
b. heat rate penalty	• restrictions in flue gas exit temperature due to limitations in catalyst operating temperature • auxiliary power, additional flue gas pressure draft	• increased fuel cost
c. reactor sootblowing and waterwashing	• influence of sootblowing frequency on deposit accumulation	• capital and operating costs of sootblowing facilities; and effluent disposal
d. catalyst life	• change in catalyst activity	• catalyst replacement
3. Environmental Impacts		
a. SO_3	• oxidation of SO_2 to SO_3 across catalyst, preheater exit concentration	• cost of SO_3 removal and disposal
b. NH_3	• carryover or breakthrough of ammonia to reactor outlet; preheater exit concentration	• cost of NH_3 removal and disposal
c. ammonium sulfate and bisulfate	• preheater exit concentration • concentration in air heater waterwash effluent	• cost of ammonium sulfate and bisulfate removal, and disposal • capital and operating costs of waterwash effluent treatment
d. interference with particulate collectibility	• influence of NH_3/SO_3 and by-products on - resistivity - physical and chemical ash characteristics	• capital cost of electrostatic precipitators - specific collecting area - ash hoppers • capital and operating cost of fabric filters - air/cloth ratio - cleaning cycle, pressure drop • ash disposal costs
e. N_2O, HCN emissions	• inlet/outlet N_2O and HCN concentration	• cost of N_2O and HCN removal
4. System-Wide Effects		
a. air heater sootblowing and waterwashing requirements	• degradation in thermal performance or pressure drop with deposit accumulation	• increased fuel costs (heat rate penalty) • capital and operating costs of sootblowing/waterwashing facilities
b. air heater operation and maintenance	• degradation in thermal performance or pressure drop with wheel basket corrosion/fouling	• increased capital and operating costs of corrosion/fouling resistant air heaters

TABLE 3

KAWASAKI HEAVY INDUSTRIES
SELECTIVE CATALYTIC REDUCTION
PROCESS-DESIGN CONDITIONS

• Flow capacity	5000 scfm (8000 Nm^3/hr)
• Catalyst quantity,space velocity	120.1 ft^3 (3.4 m^3), 2500 hr^{-1}
• Flue gas temperature	680°F (360°C)
• Fly ash loading	3.3 gr/scf (7.5gm/Nm^3)
• Flue gas composition	

Specie	Volume Concentration
O_2	5%
NO_x	600 ppm
SO_x	350 ppm
CO_2	11%
H_2O	9%
CO	15%

- NO_x removal - 90% of inlet for design flue gas conditions
  - $NH_3/NO_x \leq 1$
  - NH_3 $\leq$10 ppm (prediction only, not guaranteed)

TABLE 4

LJUNGSTROM AIR HEATER DESIGN SPECIFICATIONS

Flue Gas	
Mass in	23,000 lb/hr
Mass out	26,700 lb/hr
Temperature in	675°F
Temperature out	300°F
Pressure drop	3.2 in H_2O
Air	
Mass in	19,850 lb/hr
Mass out	16,150 lb/hr
Temperature in	85°F
Temperature out	618°F'
Pressure drop	1.3 in H_2O

Controlling Parameter	*Continuous On-Line Measurements and Processing Required*	*EPRI Experience to Date*
NH_3/NO_x	1. Flue gas flow rate and temperature 2. NO_x concentration 3. Ammonia mass flow	Limited
NO_x removal	1, 2	None
Residual NH_3	4. NH_3 flue gas concentration	None
NH_3/NO_x, maximum limited by NH_3	1, 2, 3 and 4	None
NO_x removal, maximum limited by NH_3	1, 2, 3 and 4	None

Table 5. Ammonia Injection Control Schemes Under Evaluation

Table 6.

TASK DEFINITION

TASK	Primary Variables	Fixed Parameters and/ or Secondary Variables
3. Baseline Characterization	• SCR operating parameters - NH_3/NO_x ratio - NO_x - O_2 - flue gas/catalyst temperatures • space velocity	• fuel type • particulate characterization • minor gas species - SO_2 - HC - CO
4. Transient Mode Operation	• control system logic • excursions in process gas - flowrate - temperature - composition	• fuel type • particulate characteristics • minor gas species
5. Influence of Minor Species	• SO_2 • CO • HC	• particulate characteristics • fuel type • SCR operating parameters
6. Influence of Particulates	• particulate mass loading • particulate size distribution • sootblowing schedule (accumulated solids)	• minor gas species • SCR operating parameters
7. Catalyst Activity	• erosion, aging of catalyst (hours of operation)	• particulate characteristics • minor gas species • SCR operating parameters
8. Air heater performance	• air heater operating parameters - pressure differential - wheel speed - inlet temperature - flue gas/combustion air flowrate - sootblowing frequency	• particulate characteristics • Sootblowing, medium and momentum

ASSESSMENT OF NO_x FLUE GAS TREATMENT TECHNOLOGY

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ABSTRACT

The Environmental Protection Agency has maintained a program to further the advancement of NO_x control by flue gas treatment technology since the early 1970's. The program consists of technology assessment studies in conjunction with small scale experimental projects. These activities have shown that 80-90% reduction of NO_x emissions by selective catalytic reduction with ammonia has been commercially demonstrated on gas- and oil-fired sources in Japan, and that such processes are ready for test application on coal-fired sources. The Japanese experience, combined with experimental projects in the U.S., should establish the technology as a viable control technique for use in tackling NO_x environmental problems in the U.S. However, some significant technical concerns need to be addressed in demonstration projects before widespread application of the technology can be recommended.

INTRODUCTION

The U.S. Environmental Protection Agency (EPA) has maintained a research and development program since the early 1970's to advance flue gas treatment technology for the control of nitrogen oxide (NO_x) emissions from stationary combustion sources. The objective of this program has been to demonstrate the technical feasibility of a 90% reduction of NO_x emissions in a cost effective, energy efficient, and environmentally sound manner. Emphasis of the program has been on control of NO_x emissions from coal-fired boilers.

This program has been maintained since technology for highly efficient NO_x reduction may be required as a control strategy for some of the environmental problems in the United States. Specifically, application of the technology may be required by regulations which address Prevention of Significant Deterioration, Visibility Protection, Acid Rain, or Nonattainment of National Ambient Air Quality Standards (NAAQS). The promulgation of a short term nitrogen dioxide (NO_2) NAAQS may increase the number of nonattainment areas for NO_2 and may increase the need for application of flue gas treatment technology (1).

To ameliorate their environmental problems, the Japanese have developed and applied NO_x flue gas treatment technology to gas- and oil-fired sources and are planning applications on coal-fired sources. They have also conducted extensive research on dry and wet processes for simultaneous control of NO_x and sulfur oxide (SO_x) emissions. Thus, a major portion of EPA's program has been to investigate Japanese technology for potential applications to coal-fired boilers in the U.S. Accordingly, this paper will include:

°Assessment of Technology in Japan

- Dry NO_x Processes
- Dry Simultaneous NO_x/SO_x Processes
- Wet NO_x and NO_x/SO_x Processes

°Assessment of Technology for Application in the U.S.

- Cost Estimates
- Energy and Environmental Impacts
- Experimental Projects

Finally, conclusions that can be drawn from these assessment activities will be presented.

ASSESSMENT OF TECHNOLOGY IN JAPAN

Japanese technology for control of NO_x and simultaneous control of NO_x and SO_x by flue gas treatment techniques is more advanced than any other country's. EPA has sponsored the publication of periodic reports and papers to facilitate the transfer of information from Japan to the United States. These documents have been mainly prepared by Jumpei Ando of Chuo University, Tokyo, Japan (2,3). Since most of the development and application of NO_x flue gas treatment technology has been in Japan, it is appropriate for any assessment to begin with the status of the technology in Japan.

DRY NO_x PROCESSES

Selective catalytic reduction (SCR) of NO_x with ammonia (NH_3) is the only process that has achieved notable success in treating combustion flue gas for 90% removal of NO_x and has progressed to the point of commercial application. SCR processes are based on the preferential reaction of NH_3 with NO_x rather than with other flue gas constituents. Since oxygen (O_2) enhances the reduction, the reactions can best be expressed as:

$$4NH_3 + 4NO + O_2 \longrightarrow 4N_2 + 6H_2O \qquad (1)$$

$$4NH_3 + 2NO_2 + O_2 \longrightarrow 3N_2 + 6H_2O \qquad (2)$$

Equation 1 represents the predominate reaction since approximately 95% of the NO_x in combustion flue gas is in the form of nitric oxide (NO). Therefore, under ideal conditions, a stoichiometric amount of NH_3 can be used to reduce NO_x to harmless molecular nitrogen (N_2) and water vapor (H_2O).

In practice, an NH_3:NO mole ratio of about 1:1 has typically reduced NO_x emissions by 90% with a residual NH_3 concentration of less than 20 ppm.

The SCR processes require a reactor, a catalyst, and an ammonia storage and injection system. Due to increased pressure drop across the SCR reactor, some increase in boiler fan capacity, or possibly an additional fan, may be necessary.

The optimum temperature for the NO_x reduction reaction without a catalyst is about 1000°C (1830°F). However, the catalyst effectively reduces the optimum reaction temperature to the 300° to 450°C (570° to 845°F) range. To obtain flue gas temperatures in this range and to avoid the requirement for large amounts of reheat, the reactor is usually located between the boiler economizer and the air preheater. A typical flow diagram is shown in Figure 1. Obviously, the reactor and catalyst are the critical elements of the process and warrant further discussion.

Catalysts with vanadium compounds were found to promote the reduction of NO_x with NH_3 and to be unaffected by the presence of SO_x. Titanium dioxide (TiO_2) was found to be an acceptable carrier, since it is resistant to attack from SO_3. Therefore, many SO_x resistant catalysts are based on TiO_2 and V_2O_5; however, constituents and concentrations of most catalysts are proprietary.

Reactor and catalyst configurations also vary with the application, primarily to accommodate the different particulate concentrations. Natural-gas-fired boilers employ SCR catalysts (as spherical pellets, cylinders, or rings) and reactor vessels (as fixed packed beds).

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However, designs for use with oil- and coal-fired boilers have to be capable of tolerating particulates (fly ash) in the flue gas stream. For these applications, a parallel flow catalyst is preferred. Parallel flow means that the gas flows straight through the open channels parallel to the catalyst surface. The particulates in the gas remain entrained while NO_x reaches the catalyst surface by turbulent convection and diffusion. An alternative to the parallel flow catalysts is the parallel passage reactor. In this design, the catalyst material is arranged in channels and held in place by a metal screen. The operating principle is similar to that of the parallel flow catalyst.

The various parallel flow catalyst shapes are shown in Figure 2. The catalyst may be a homogeneous material or may be composed of an active material coated on the walls of a metallic or ceramic carrier. The parallel flow catalysts are normally manufactured in a unit cell configuration about 1 m^3 as shown in Figure 3. The cells are stacked in banks in the reactor as shown in Figure 4.

Even though much progress has been made in catalysts and reactor design, some problems still remain. The catalysts may not be resistant to all contaminants in flue gas or be able to tolerate high particulate loadings. In addition, fine particulates, smaller than about 1 μm, may blind the catalyst surface. Long term operation without catalyst plugging or catalyst erosion needs to be demonstrated for coal-fired applications. Catalyst life also needs to be extended from the current guarantees of 1 to 2 years for applications with SO_x and particulates in the gas stream.

One of the major concerns with SCR processes is the formation of solid ammonium sulfate [$(NH_4)_2SO_4$] and liquid ammonium bisulfate (NH_4HSO_4) downstream of the reactor. The formation conditions are difficult to avoid since some unreacted NH_3 from a SCR system and some SO_3 from combustion of sulfur-containing fuels is expected. The biggest problem seems to be deposition of $(NH_4)_2SO_4$ and NH_4HSO_4 on the air preheater. These compounds are corrosive and can form deposits which plug the air preheater. Air preheater problems were found to be most severe on high-sulfur oil-fired applications and coal-fired units which employ a hot electrostatic precipitator (ESP) ahead of the NO_x reactor and the air preheater. Systems which accept full particulate charge of a coal-fired boiler through the air preheater should have less difficulty with pluggage of the air preheater. In these systems, the fly ash apparently scours the surface of the air preheater to remove any deposits, or the ammonium compounds deposit on the fly ash and are carried through the air preheater by the fly ash. However, increased soot blowing, from both the hot and cold sides, and water washing of the air preheaters appear necessary for most applications. The wash water from cleaning air preheaters as well as purge streams from a flue gas desulfurization (FGD) unit may require treatment to remove NH_3 before being discharged. Modifications to the conventional air preheater design are also being developed to address the ammonium sulfate problem; however, these units have not been applied on full scale systems as yet.

Other concerns and potential problems include: emission of NH_3 and NH_3 compounds; causing or increasing the emission of undesirable compounds such as SO_3; affecting the performance of downstream pollution control equipment such as FGD processes, ESPs, and baghouses; lack of proven NH_3 analytical control systems; sensitivity of the process to temperature changes due to boiler load swings; disposal or reclamation of spent catalysts in an environmentally acceptable manner; and reliability of the process and its effect on the boiler system's availability.

Despite these potential problem areas and uncertainties, the processes have been successfully installed and operated in Japan on gas- and oil-fired boilers, and coal-fired units are being constructed. The companies listed in Table I were visited by a team of EPA, Tennessee Valley Authority, and Radian Corporation personnel in March 1980 to survey the status of the technology in Japan. The gas- and oil-fired operating units reported very high reliability and the capability to overcome operational difficulties. In general, no labor was added to operate a SCR system on retrofit applications, and new systems do not plan additional personnel for the NO_x system. Generally, no maintenance was reported other than cleaning the air preheater during annual shutdowns. The control systems are usually fairly simple and employ a feed-forward control system based on inlet NO_x concentration with fine tuning supplied by feedback of the outlet NO_x concentration. Most gas- and oil-fired applications also utilized an outlet NH_3 emissions monitor; however, an NH_3 emissions monitor has not been perfected for coal-fired applications. NH_3 emissions were reported to be very low (0-5 ppm) in most cases. NH_3 emission limitations are not prescribed by governmental agencies, but low NH_3 emissions are deemed essential to minimizing problems with downstream equipment. No problems were noted with NH_3 fuming or emissions of cyanide, nitrates, nitrosoamines, or other harmful species.

The NO_x emission rates were very low when SCR units were employed. As shown in Table I, gas-fired units were controlled to about 10 ppm of NO_x and oil-fired units to 15-35 ppm. Further, coal-fired units are being constructed which will demonstrate control to 40-80 ppm of NO_x. It is important to note that the Japanese seem to prefer 80% NO_x removal as the optimum control level since it minimizes capital and operating costs as well as energy and environmental impacts while maximizing operability, reliability, and system life.

The coal-fired applications of SCR technology in Japan are of particular interest to U.S. personnel since coal-fired sources represent a major NO_x control problem in the U.S. Four SCR systems for coal-fired boilers were under construction in Japan and are shown in Table I. Further, construction of additional coal-fired boilers are being planned and SCR units are expected to be installed on most of these units. Therefore, the Japanese experience should provide valuable information for many years for U.S. personnel interested in NO_x control.

The SCR units were installed to meet local governmental NO_x emission limitations and are not necessary to comply with the Japanese national standard. Compliance with local and national environmental agreements is achieved through a sincere cooperative spirit that exists between industry and government to solve the country's pollution problems. Local governments maintain a very comprehensive and extensive instrumentation system to monitor ambient pollutant concentrations and emission rates from major emitters. Although the local governments have no enforcement authority, violations are rarely, if ever, detected since the companies do not want to jeopardize the "good will" that exists between the industrial, governmental, and public sectors.

In summary, it should be emphasized that SCR technology is being successfully operated in Japan on large scale gas- and oil-fired sources and that large scale applications to coal-fired sources are under construction with equal success anticipated.

DRY SIMULTANEOUS NO_x/SO_x PROCESSES

The Shell Flue Gas Treating process is a unique variation of SCR technology that can simultaneously remove both NO_x and SO_x from combustion flue gas. The process uses copper oxide (CuO) supported on stabilized alumina placed in two or more parallel-passage reactors. The reactions, which characterize process operation, can be expressed as:

Acceptance

$$CuO + 1/2\ O_2 + SO_2 \longrightarrow CuSO_4 \qquad (3)$$

$$4NO + 4NH_3 + O_2 \xrightarrow{CuSO_4} 4N_2 + 6H_2O \qquad (4)$$

Regeneration

$$CuSO_4 + 2H_2 \longrightarrow Cu + SO_2 + 2H_2O \qquad (5)$$

$$Cu + 1/2\ O_2 \longrightarrow CuO \qquad (6)$$

Flue gas is introduced at 385°C (725°F) into one of the reactors where the SO_2 reacts with CuO to form copper sulfate ($CuSO_4$). The $CuSO_4$ and, to a lesser extent, the CuO act as catalysts in the reduction of NO_x with NH_3. When the reactor is saturated with $CuSO_4$, flue gas is switched to a fresh reactor for acceptance of the flue gas, and the spent reactor is regenerated. In the regeneration cycle, hydrogen (H_2) is used to reduce the $CuSO_4$ to copper (Cu), yielding a SO_2 stream of sufficient concentration for conversion to sulfur or sulfuric acid. The Cu in the reactor is oxidized, preparing the reactor for acceptance of the flue gas again. Between acceptance and regeneration, steam is injected into the reactor to purge the remaining flue gas or H_2 to eliminate any possibility of combustion. The process can also be operated in the NO_x-only mode by eliminating the regeneration cycle, or in the SO_x-only mode by eliminating the NH_3 injection.

The process has been installed in Japan on a heavy-oil-fired boiler treating 120,000 Nm^3/hr of flue gas. The unit has demonstrated 90% SO_2 removal and 70% NO_x removal. UOP Process Division is the licensor of the process in the United States.

WET NO_x AND NO_x/SO_x PROCESSES

The wet NO_x and simultaneous NO_x/SO_x processes developed to date cannot compete economically with dry processes for removal of NO_x from combustion flue gas. This is primarily due to the complexity, limited applicability, and water pollution problems associated with the wet processes.

ASSESSMENT OF TECHNOLOGY FOR APPLICATION IN THE U.S.

Since NO_x SCR systems have been successfully installed and operated in Japan on gas- and oil-fired sources, it is expected that equal success could be obtained in the U.S. However, coal-fired boilers in the U.S. are the primary concern for control of NO_x from stationary combustion sources. Therefore, EPA has focused on technology assessment studies to determine cost estimates, energy requirements, and environmental impacts of applying the technology on coal-fired boilers in the U.S. In parallel with the technology assessment studies, small scale experimental projects have been undertaken to demonstrate the performance of the technology.

COST ESTIMATES

Preliminary cost estimates for application of SCR processes to a coal-fired utility boiler were made by the Tennessee Valley Authority through an interagency agreement with EPA. (This study was cosponsored by the Electric Power Research Institute.) The basis for this estimate was a new, 500 MW boiler firing Eastern bituminous coal with a heating value of 24.4 MJ/kg (10,500 Btu/lb), a sulfur content of 3.5%, and an ash content of 16%. The study assumed operation of the boiler for 7000 hr/yr and the SCR process for 90% reduction of NO_x. The estimates were based on a capital investment in mid-1979 and an annual revenue requirement in mid-1980. Results of that study, summarized in Table II, indicate that the capital costs of SCR processes will be about $42/kW or $9.8/acfm and that annual revenue requirements will be about 2.7 mills/kWh. (Since the volume of gas treated is the dominating parameter in determining the cost of installing an SCR system, the $/acfm factor may be the most meaningful in determining representative costs of firing various coals in various sized plants.) The study also indicated that the dry simultaneous NO_x/SO_x process would be competitive with these costs if the cost of a FGD process for SO_2 control were added to the cost of the NO_x SCR process, but that the cost of wet simultaneous NO_x/SO_x processes would be significantly higher.

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The cost of applying NO_x SCR technology to industrial boilers was estimated in a study for EPA by Radian Corporation (5). It was found that the fuel fired, boiler size, and design NO_x removal had significant effects on the system cost. The results of this study are summarized in Table III.

ENERGY AND ENVIRONMENTAL IMPACTS

The SCR processes are projected to require about 0.2% of the boiler capacity. This assumes that flue gas reheat will not be required and does not include an energy requirement represented by raw materials (4).

The major concerns from an environmental impact viewpoint appear to be emissions and discharge of NH_3 and NH_3 compounds and disposal of spent catalysts. Operational techniques to limit NH_3 emissions and methods for regenerating or reclaiming the catalysts are under development to minimize problems in this regard (5).

EXPERIMENTAL PROJECTS

To determine the actual performance and cost of applying SCR processes in the U.S., several demonstration projects have been planned as summarized in Table IV. Relative to the EPA pilot plant projects, the basic objective is to demonstrate the feasibility of the processes for 90% control of NO_x or NO_x/SO_x emissions from a coal-fired source.

The projects were divided into four phases:

I - Design
II - Procurement and erection
III - Startup, debugging, and optimization
IV - Long term operation and assessment.

Phase IV began in June 1980 for both projects. Therefore, it is not possible to provide results of the projects in this paper. However, it should be noted that numerous problems have been encountered in operating the pilot plants. One major problem area has been sampling and analytical techniques for NO, NO_2, NO_x, SO_2, SO_3, and NH_3. Successes and failures of the pilot plants will be reported in full in the final project reports. Since papers on each project or process listed in Table IV will be presented at this Symposium, further details will not be presented in this paper.

If the coal-fired pilot plants and oil-fired demonstration scale plants are successful, then a coal-fired prototype or demonstration scale plant (10-100 MW) will be desirable. It is conceivable that such a plant could be built in conjunction with a new or expanding facility in an area with nonattainment, prevention of significant deterioration, visibility, or other environmental constraints. In this manner, the environmental impact of the facility could be minimized, and SCR technology for NO_x control could be commercially demonstrated.

CONCLUSIONS

The following conclusions can be drawn from this assessment of NO_x flue gas treatment technology.

1. NO_x flue gas treatment processes, based on selective catalytic reduction (SCR) of NO_x with NH_3, have been successfully demonstrated on commercial-scale gas- and oil-fired sources in Japan for $\geq$80% NO_x removal.

2. SCR systems are being installed on commercial-scale coal-fired sources in Japan which will demonstrate the viability of the technology for coal-fired sources.

3. SCR systems are being evaluated on pilot-scale coal-fired sources and on a demonstration-scale oil-fired source in the U.S. for 90% NO_x control. One of the pilot plants is evaluating dry simultaneous NO_x/SO_x control. If these applications are successful, evaluation on a prototype-scale coal-fired facility will be desirable.

4. Although significant progress has been made in developing and applying the technology, several problem areas remain to be resolved (i.e., impact on downstream equipment, long term performance, and reliability).

5. Experimental projects, in conjunction with technology asssessment studies, will enable a determination of the feasibility of applying NO_x SCR processes in the U.S.

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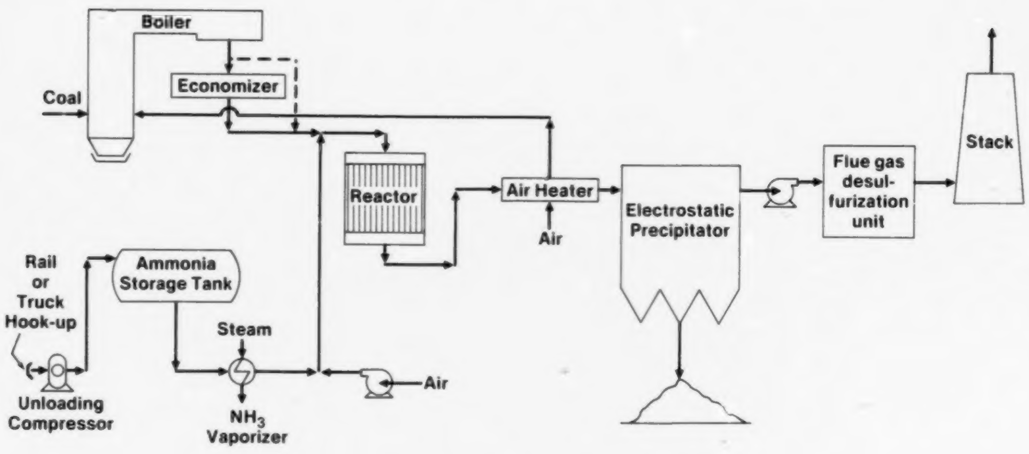
Boiler
Economizer
Coal
Reactor
Air Heater
Air
Electrostatic Precipitator
Flue gas desul-furization unit
Stack
Rail or Truck Hook-up
Ammonia Storage Tank
Steam
Air
Unloading Compressor
NH_3 Vaporizer

Figure 1. Typical flowsheet for NO_x selective catalytic reduction processes (4).

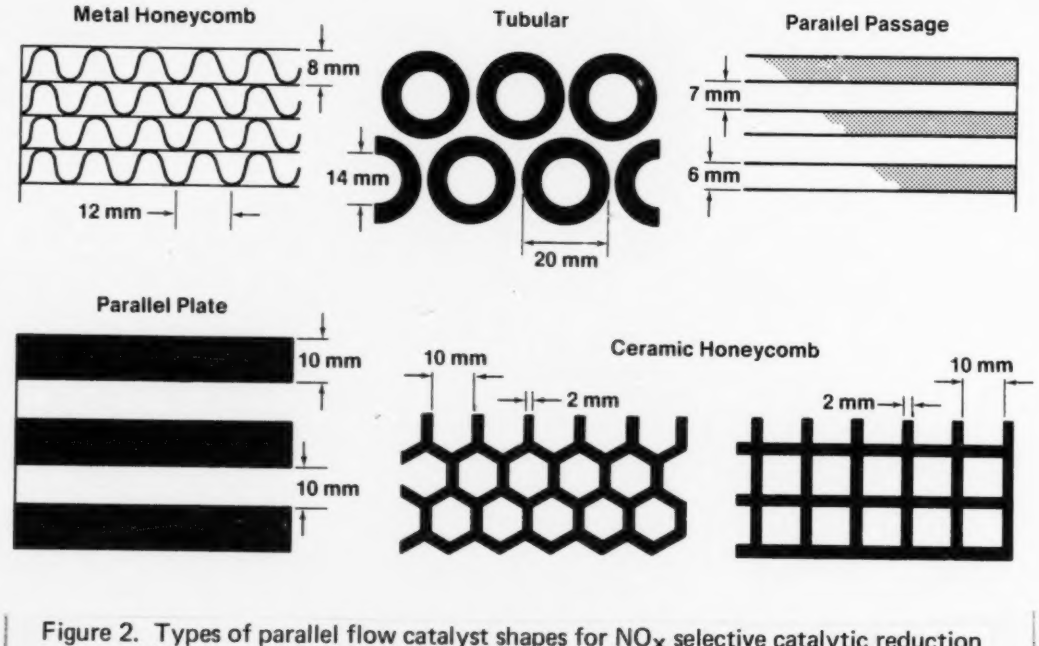
Metal Honeycomb
8 mm
12 mm
Tubular
14 mm
20 mm
Parallel Passage
7 mm
6 mm
Parallel Plate
10 mm
10 mm
Ceramic Honeycomb
10 mm
2 mm
2 mm
10 mm

Figure 2. Types of parallel flow catalyst shapes for NO_x selective catalytic reduction processes (3).

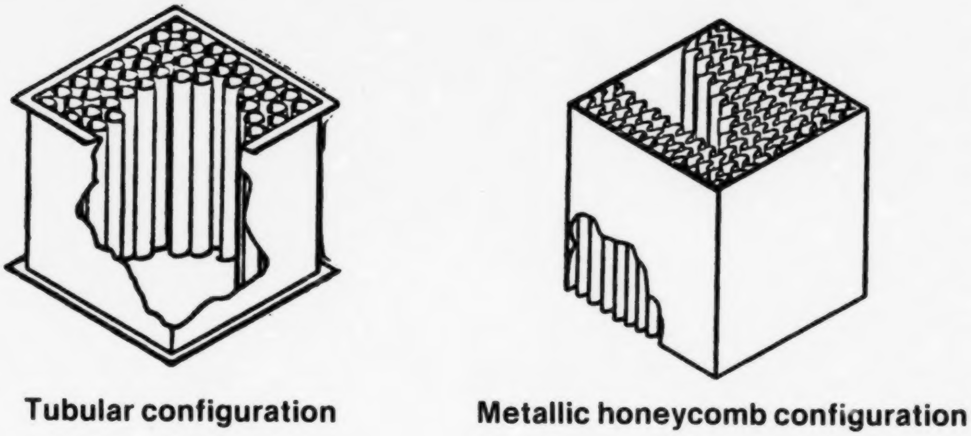
Tubular configuration
Metallic honeycomb configuration

Figure 3. Unit cells of parallel flow catalysts for NO_x selective catalytic reduction processes (3).

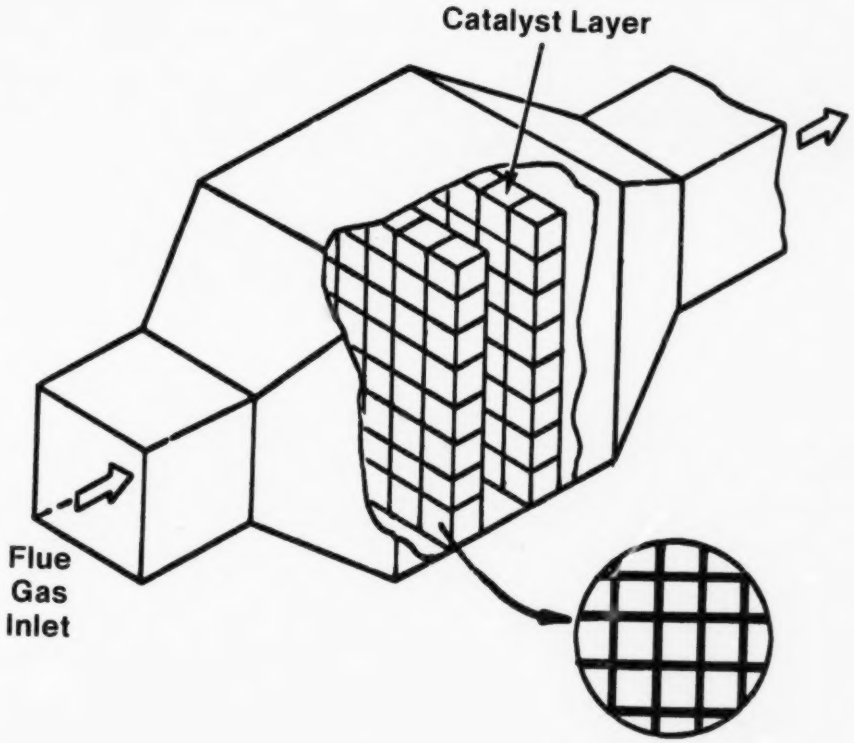
Catalyst Layer
Flue
Gas
Inlet

Figure 4. Parallel flow reactor for NO_x selective catalytic reduction processes (3).

TABLE I. SELECTED INSTALLATIONS WITH NO_x SELECTIVE CATALYTIC REDUCTION SYSTEMS IN JAPAN[a]

		Boiler			NO_x SCR System				
Company	Station	No.	Fuel	Size (MW)	Size (MW)	NO_x Removal (%)	Outlet NO_x (ppm)	Vendor[b]	Startup
Chubu Electric	Chita	5	Gas	700	700	80	8-10	B-H	Mar. 1978
		6	Gas	700	700	80	8-10	B-H	Apr. 1978
Chugoku Electric	Kudamatsu	4	Oil	700	700	80	20	MHI	Mar. 1980
		2	Oil	375	375	80	32	IHI	Apr. 1979
		3	Oil	700	700	80	26	IHI	Sep. 1979
Fuji Oil	Sodegara	7	Oil	40	40	87	15	MHI	Jan. 1978
		—	CO & Oil	10	10	92	15	JGC	Jul. 1976
Chugoku Electric	Shimonoseki	1	Coal	175	175	50	250	MHI	Apr. 1980
Hokkaido Electric	Tomakomai	1	Coal	350	90	>80	<40	B-H	Oct. 1980
Electric Power Development Co.	Takehara	1	Coal	250	125	80	80	B-H	Jun. 1981
		1	Coal	250	125	80	80	KHI	Jun. 1981

a) These installations were visited by a team of EPA, TVA, and Radian personnel in March 1980.

b) B-H Babcock-Hitachi
MHI Mitsubishi Heavy Industries
IHI Ishikawajima-Harima Heavy Industries
JGC JGC Corporation
KHI Kawasaki Heavy Industries

TABLE II. ESTIMATED COSTS FOR EMISSION CONTROL SYSTEMS FOR UTILITY BOILERS

Process Types	Capital Costs ($/kW)				Annualized Costs (mills/kWh)			
	NO_x	SO_2	PM	Total	NO_x	SO_2	PM	Total
SCR, FGD, ESP	42	101	22	165	2.7	4.2	0.7	7.6
Dry Simultaneous NO_x/SO_x	←134→		29	163	←6.4→		0.9	7.3
Wet Simultaneous NO_x/SO_x/PM	←200→			200	←11.3→			11.3

Basis for the Estimate:

NO_x Control System	Selective Catalytic Reduction (SCR) with NH_3
SO_2 Control System	Limestone Flue Gas Desulfurization (FGD)
Particulate Matter (PM) Control System	Electrostatic Precipitator (ESP) for dry systems; wet scrubber for wet simultaneous systems
NO_x Removal Efficiency	90%
SO_2 Removal Efficiency	90%
PM Removal Efficiency	99.5%
Boiler Size	500 MW, new
Fuel	Coal
Heating value	24.4 MJ/kg (10,500 Btu/lb)
Sulfur content	3.5%
Ash content	16%
Operation	7000 hr/yr
Capital Investment	mid-1979
Annual Revenue Requirement	mid-1980
SCR Processes	Average of UOP-Shell, Hitachi Zosen, Kurabo process costs
Dry Simultaneous NO_x/SO_x	UOP-Shell
Wet Simultaneous NO_x/SO_x/PM	Average of Moretana Calcium and Asahi ClO_2 process costs

Source: Maxwell, Burnett, and Faucett (4).

TABLE III. ESTIMATED COST OF NO_x SELECTIVE CATALYTIC REDUCTION SYSTEMS FOR INDUSTRIAL BOILERS

Fuel	Rating		Capital Costs $			Annual Costs $		
	(MJ/s)	(MBtu/hr)	70% Control	80% Control	90% Control	70% Control	80% Control	90% Control
Coal	8.8	30	181,900	213,100	249,600	104,200	117,500	133,900
(Parallel Flow)	22.0	75	298,900	349,500	413,300	157,600	170,700	196,500
	44.0	150	451,500	534,600	633,000	216,600	251,600	292,300
	58.6	200	531,900	632,600	752,400	254,200	298,800	351,300
Residual Oil	8.8	30	158,100	179,100	202,900	96,100	102,000	108,200
(Parallel Flow)	44.0	150	378,200	436,200	503,100	181,180	200,900	222,860
Residual Oil	8.8	30	144,600	154,700	166,700	120,400	125,100	129,900
(Moving Bed)	44.0	150	259,700	300,200	347,000	167,700	184,800	203,600
Distillate Oil	4.4	15	88,200	93,600	99,400	63,600	65,500	67,400
(Fixed Packed Bed)	44.0	150	223,800	261,500	305,600	137,200	155,300	175,900
Natural Gas	4.4	15	92,800	96,600	100,500	64,400	66,000	67,600
(Fixed Packed Bed)	44.0	150	223,700	262,000	306,800	129,400	150,400	174,700

Basis for Estimate:	
Cost Basis	Mid-1978
Load Factors	
Coal	0.60
Residual Oil	0.55
Distillate Oil	0.45
Natural Gas	0.45
Coal	Low Sulfur Western

Source: Jones and Johnson (5).

TABLE IV. PLANNED DEMONSTRATION PROJECTS OF NO_x SELECTIVE CATALYTIC REDUCTION TECHNOLOGY IN THE U.S.

Sponsor	Contractor	Location	Size (MWe)	Fuel	Planned NO_x Reduction	Startup
Environmental Protection Agency	Hitachi Zosen	Georgia Power Company	0.5	Coal	90%	Fall 1979
Environmental Protection Agency	UOP Process Division	Tampa Electric Company	0.5	Coal	90%(a)	Fall 1979
Electric Power Research Institute	Kawasaki Heavy Industries	Public Service Company of Colorado Arapahoe Station	2.5	Coal	90%	Winter 1980
Southern California Edison(b)	Kawasaki Heavy Industries	Southern California Edison Huntington Beach Station	100	Oil	90%	Fall 1981

a) 90% Reduction of SO_2 also planned by the dry, simultaneous NO_x/SO_x process.

b) Required to meet regulations of the California Air Resources Board.

DEVELOPMENT OF FLUE GAS TREATMENT IN JAPAN

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ABSTRACT

The Electric Power Development Co., Ltd. (EPDC) has been executing a research and development program on selective catalytic reduction (SCR) systems through joint research with the equipment manufacturers since 1975.

As a result of this R&D program, EPDC believes there is a strong prospect for commercialization of the Low Dust SCR System (LDSS) for coal-fired power plants. At present, EPDC is constructing demonstration test equipment for such a system at the Takehara Thermal Power Station No. 1 Unit (250 MW coal-fired) and plans to construct full scale commercial SCR equipment at the Takehara Thermal Station No. 3 Unit (700 MW coal-fired).

Alternatively, the High Dust SCR System (HDSS) is also a candidate for commercialization, requiring only the establishment of NH_3 removal technology for ash collected by a cold-side electrostatic precipitator.

This report describes the results of the R&D program executed by EPDC concerning the SCR and air preheater problems, overall flue gas treatment technology for coal-fired boilers, and the outline of Takehara's SCR Systems.

Section 1

INTRODUCTION

The oil shortage which gripped the world in the fall of 1973 triggered a series of oil supply uncertainties in the years that followed, emphasizing the importance of renewed efforts to increase the utilization of coal.

Coal-fired thermal power generation will be the major focus of this shifted emphasis to coal. A coal-fired thermal power plant, however, must be well controlled in order to satisfy all environmental protection requirements. Otherwise, the utility may encounter resistance from governmental agencies or local residents, resulting in failure to obtain a site.

A thermal power plant using coal as fuel, compared to one using other fossil fuels, emits larger quantities of dust (particulates), SO_x, and NO_x from its boiler, necessitating special measures to hold down these emissions. Such controls include auxiliary equipment for dust collection, desulfurization and denitration, including combustion modifications.

Electric Power Development Co. (EPDC), in cooperation with certain Japanese manufacturers, has been conducting research and development on technologies for the control of emissions from thermal coal-fired power generation plants. Currently, assured of successful commercialization of boiler denitration technology, EPDC is installing SCR demonstration equipment on a coal-fired thermal power plant. In addition, EPDC has just started construction of a 700 MW coal-fired thermal power plant featuring similar SCR equipment.

In this paper, we introduce several problems encountered in placing denitration equipment in commercial operation together with remedial measures, as well as details about denitration performance, ammonia injection control, and other parameters. Also, we will outline our progress in the development of emission control technologies for other emission from a coal-fired thermal power plant.

Section 2

GENERAL DESCRIPTION

INSTALLATION OF SCR EQUIPMENT FOR ELECTRIC POWER UTILITIES IN JAPAN

Table 1 shows a list of Selective Catalytic Reduction (SCR) equipment installed or in the planning stage by Japanese electric power utilities. Of the plant sites shown in the table, two are operating with SCR equipment for coal-fired power generation, and another two are being installed with similar equipment. Of the coal-fired power generation plants listed in the table, the Tomatoazuma Power Plant of the Hokkaido Electric Power Co., Inc. is provided with SCR equipment designed on a 1/4-scale capacity; the Shimonoseki Power Plant of the Chugoku Electric Power Co., Inc. with equipment designed for full capacity.

EPDC has been commissioned by the government to construct SCR equipment designed for the full capacity of the No. 1 boiler (250 MW) of the Takehara Thermal Power Plant. This demonstration SCR equipment is designed to operate at 80 percent or more NO_x removal. In addition, EPDC plans to provide the recently constructed No. 3 thermal power generation boiler (700 MW) with similar SCR equipment designed to operate at 80 percent or more NO_x removal. These plans should be considered representative of future programs for construction of coal-fired thermal power generation plants.

EVALUATION OF DENITRATION PROCESS

Studies of the denitration process directed to coal-fired thermal systems have evaluated the following:

- economic efficiency
- performance

- reliability
- secondary environmental emissions
- retrofit capability

Japanese NO_x regulations are stringent; thus it was important that the $DeNO_x$ Process be quickly evaluated, and brought to commercialization in a short period of time. As a result of a comprehensive analysis, EPDC concluded that selective catalytic reduction (SCR) using ammonia was the most suitable process for near-term application.

The comparison of results between SCR and selective non-catalytic reduction (SNR), including the practical use of wet $DeSO_x$, is shown in Table II.

CATALYST COMPARISON BY TYPES OF FUEL

Research and development of the SCR process for oil-fired boilers has been executed since 1973, and for coal-fired boilers since 1975. Process development for power plants has focused on the development of a suitable catalyst. Coal combustion results in large quantities of dust and SO_x becoming entrained in the flue gas,thus, catalyst development for coal combustion has special problems such as deterioration and plugging. Table III shows catalyst features for different types of fuel.

DEVELOPMENT OF SCR FOR COAL-FIRED BOILERS

It is not possible to apply the results of SCR development from LNG and oil-fired boilers to coal-fired boilers because of differences in flue gas components; thus there is a need to assess the practicality of several catalysts for coal-fired boilers.

Among power utilities in Japan, EPDC, Tokyo Electric Power Company (TEPCO), and Hokkaido Electric Power Company (HEPCO), have individually tested the SCR process through pilot tests at their own power plants. Table IV shows the outline of the pilot tests.

Section 3

DEVELOPMENT OF AN SCR SYSTEM

EPDC, in developing the SCR system has identified the following needs:

- catalyst life must be greater than one year
- the system must be appplicable to both high and low dust loading
- continuous measuring instruments for leak ammonia and countermeasures for leak ammonia influence on downstream equipment are required.
- recycle or reutilization of spent catalyst

DEVELOPMENT OF CATALYST LIFE OVER ONE YEAR

For practical use of SCR, the following targets must be satisfied during the one year of pilot tests:

- maintaining more than 80% efficiency
- leak ammonia should be less than 5 ppm

The following discussion details how these targets have been met by the 5 joint researchers of EPDC.

Catalysts with 2 years of life have been promised by the manufacturers, although no experiments have yet been performed testing catalysts contінously for over 2 years. The catalyst cost is relatively high, comprising 40-60% of the total SCR equipment cost. Whether catalyst life is 1 or 2 years will be a significant influence on electricity cost; therefore, it is important to develop long life catalysts. At this time no assessment of catalyst life is possible as pilot tests of sufficient duration have not been conducted. However, the following catalyst performance test results suggest extended life is probable:

- Performance decreased slightly from initial values but stabilized after 500 - 3000 hours.

- Significant performance drop was observed using catalysts without treatment for SO_x poisoning; treated catalysts performed slightly better.

Catalyst performance drop is thought to be caused by the following:

(a) plugging of catalyst pores by NH_3/SO_3 compounds

(b) plugging of catalyst layer and catalyst pores by dust

(c) catalyst erosion by dust

(d) permanent posion by fluorine, chlorine and calcium

Our experience from pilot and laboratory tests indicates neither significant plugging of the catalyst layer and pores nor dust erosion. Thus, poisoning by calcium or similar elements is suggested, making it necessary to check the fuel calcium content before applying SCR to power plants.

Catalyst damage caused by temperature variation associated with daily load changes or start-up and shut-down is of concern and also will have to be evaluated through long term testing.

SCR SYSTEM

Two systems for SCR have been developed, a Low Dust SCR System (LDSS), which removes NO_x after removing dust (by a hot-side electrostatic precipitator), and a High Dust SCR System (HDSS), which removes NO_x before removing dust (by a cold-side electrostatic precipitator). These systems are shown in Figure 1.

Low Dust System vs. High Dust System

The low dust and high dust SCR systems each have their own merit and demerits. For comparison of these systems, it is necessary to examine not only the SCR equipment, but also the entire coal-fired thermal power generation system, in accordance with Table V.

Before details about the results of these studies are discussed, it must be pointed out that the choice between HDSS and LDSS, aside from the technical details involved, depends on a combination of several factors, such as environmental goals, relevant regulatory standards, and the goals of individual electric power corporations. Under these circumstances, it is yet to be determined which of these two systems will be adopted in Japan.

Overall Thermal Power Plant System

The following lists several problems characteristic of LDSS and HDSS SCR equipment:

- Compared with HDSS, LSDD offers a greater potential for the air-preheater being blocked, necessitating the conventional type air preheater to be replaced with a special one intended for SCR.

- The LDSS, in which a precipitator (hot side) is installed upstream of SCR, will not contaminate collected ash with NH_3; the HDSS, in which a precipitator (cold-side) is installed downstream of SCR, will collect NH_3 ash contaminated.

To use NH_3 contaminated ash as landfill material could require certain countermeasures, depending on the quantity of NH_3 entrained and the specific diposal circumstances.

The Table VI shows the results of examination with reference to other items.

Precipitator

A major problem associated with the HDSS system concerns the possibility of leak ammonia mingling with fly ash, leading to a possible decline in quality of the fly ash. It seems technically possible to minimize the leak ammonia to the low levels of 5 ppm or less, which, in our opinion, will not cause any deterioration in fly ash quality.

SCR Equipment

Table VII shows the results of comparing the HDSS and LDSS. Of particular interest is the fact that both these systems are free from the problem of the catalyst layer being blocked, provided special consideration is given to the catalyst shape and the gas flow speed. LDSS also appears more tolerant to surface masking by fine particulates than the HDSS. The latter phenomenon was witnessed using a scanning electron microscope to examine the catalyst surfaces. Thus, LDSS requires the installation of a soot blower.

However, EPDC, in cooperation with certain Japaneses manufacturing firms, has developed a catalyst capable of maintaining a specified NO_x removal capacity for a period of one year without a soot blower. Thus, EPDC presently has no intention of using a soot blower at this time.

Desulfurization Equipment

Another problem characteristic to SCR lies in the possibility of the ammonia entering the desulfurization system and becoming concentrated in the rejected solid waste. In such a case, it is necessary to install additional NH_3 removal equipment for the LDSS system. However, the HDSS allows the leak ammonia to be almost fully adsorbed by the fly ash and removed through the cold-side ESP, thus eliminating the need to install equipment for significant removal of NH_3.

The Table IX shows the results of comparing LDSS and HDSS.

LEAK NH_3 FROM SCR EQUIPMENT - POTENTIAL IMPACTS AND COUNTERMEASURES

The following discussion is directed to the problems attributable to leak NH_3 from SCR equipment. EPDC has been executing R&D programs to establish countermeasures for:

- Concentration of ammonia in Wet $DeSO_x$ (HDSS and LDSS).
- Plugging of the air preheater (HDSS and LDSS).
- Fly ash contaminated with NH_3 and collected by electrostatic precipitators (Refer to Figure 2).

Removal of NH_3/N in Waste From Wet $DeSO_x$

EPDC has executed R&D programs in cooperation with Japanese manufacturers for removing NH_3 and N concentrated in Wet $DeSO_x$ System. This R&D was successful, resulting in total waste water nitrogen (N) less than 10 ppm.

Presently, EPDC is constructing N and NH_3 removal processes for the SCR demonstration test equipment at Takehara power station No. 1 unit. Specifications of the process are as follows:

• Capacity	1,000 m^3/day
• Quantity of NH_4^+	198 Kg-N/day
• Total N at Outlet	10 ppm and below

Plugging and Corrosion of the Air Preheater

Plugging and corrosion of air preheaters has been addressed since the initial phases of SCR development. EPDC has been executing R&D programs for this problem in cooperation with air preheater manufacturers, using Ljungstrom or Rothemulle pilot test equipment. To date, results have been obtained on the Ljungstrom equipment. For the HDSS case, plugging was not observed, because of the self cleaning and scouring action of the dust. For the LDSS case, this scouring action does not exist; thus some countermeasures to plugging are necessary. Countermeasures investigated shown in Table XII.

Plugging tendencies of air preheaters with the LDSS system were confirmed in 1976. Work since then has not identified the plugging problems even after one year of testing. Additional tests to define countermeasures for LDSS air preheater plugging and the effects of such countermeasures have been conducted and the results are shown in Figure 5.

Major countermeasures being evaluated for plugging include:

- Pressure, steam quantity and time of soot blowing, and improvement of the soot blower design
- Change from DF to NF type of inter-element, and a one piece element for intermediate and cold segments.

The air preheater for LDSS applications is larger than that for HDSS and therefore less economical. Future plans call for continued development of a new soot-blower, with pilot tests in near future. For the Rothemuhle air preheater, detailed results are described in IHI's paper in this symposium proceedings.

REGENERATION OF THE CATALYST

EPDC has been executing an R&D project in cooperation with Kawasaki Heavy Industries concerning regeneration of spent catalyst. The objective is to imporve catalyst utilization and thus the economics of the SCR system. Four methods of regeneration have been evaluated, based on (1) steam heating, (2) purging with high temperature flue gas, (3) water cleaning, and (4) water cleaning using sonic impulses. Each of these methods was applied to catalysts forced to deteriorate in pilot test plants. The results have confirmed that reactivation of the deteriorated catalyst to the initial performance level is possible.

Tests are currently being conducted with the reactivated catalyst in the pilot plant, and no deterioration of catalyst performance has been observed after 3,000 hours.

SCR PERFORMANCE

EPDC has specified 80% NO_x removal as design conditions for performance. However, we are targeting for levels as high as 90% and above while simultaneously maintaining a maximum of 5 ppm leak NH_3 by employing changes in catalyst volume and NH_3/NO_x ratio. An increase in NO_x removal efficiency from 80% to 90% under these conditions requires an increase in catalyst volume of 30-50%. Thus, high removal efficiencies require greater capital expenditure, and necessitate higher draft losses, and higher SO_2 to SO_3, conversion rates. This trend is exhibited in Figure 6 which shows the relationship between space velocity (SV) and NO_x removal efficiency. These data indicate that the increase in catalyst volume, required from to move from 80% removal (.8 NH_3/NO_x) to 90% removal (.9 NH_3/NO_x) is approximately 50% (5,400 + 3,800 h^{-1}).

A systemwide analysis of SCR operation at 80% removal efficiency will be conducted on the demonstration test equipment of Takahara Unit No. 1. Depending on these results, a similar analysis of systemwide operation at 90% removal efficiency may be conducted.

DEVELOPMENT OF A CONTINUOUS NH_3 ANALYZER

Continuous analysis of NH_3 in flue gas is required to (a) precisely measure the concentration of injected NH_3, and (b) monitor residual NH_3 to indicate the potential for air heater problems and upsets in SCR operation. Continuous analysis for NH_3 on both a conductivity and infrared gas basis has been explored. However, these methods appear to be unfavorable for use as process-monitoring devices due to unacceptable response time, limits of detection, and stability.

Recently, a method employing the conversion of NH_3 to NO was developed. This method infers the NH_3 concentration by measuring the difference in the sample before and after converison of NH_3 to NO. Difficulties with this technique include (1) a large measurement error due to high background NO levels, and (2) short lifetimes of the NH_3 to NO converter.

Conditions are most favorable for this method when the gas sample line length is relatively short. Otherwise, ammonia is adsorbed in the inner surface of the sampling line at low temperature, or lost by the chemical reaction between NH_3 and SO_3. For coal-fired flue gas, this approach appears capable of NH_3 measurement with dust in the sample gas while requiring acceptable levels of maintenance.

EPDC has also evaluated a continuous NH_3 analyzer developed by Anritsu Electric Co., Ltd. based on the self-modulated derivative spectrometry of the ultraviolet absorption of NH_3 molecules. The NH_3 analyzer has been used since 1977 in SCR tests executed by EPDC and the Hitachi Shipbulding and Engineering Co., Ltd. (HZ).

The principle of this NH_3 analyzer is based on the following:

1. Ammonia molecules have a periodic absorption spectrum (shown in Figure 7)
2. The light transmitted into the ammonia gas also has a periodic spectrum (shown in Figure 8a).
3. The wavelength is modulated around the intensity minimum (Figure 8b).
4. Intensity-Modulated Light Single (IMLS) is obtained (Figure 8c) and the amplitude of IMLS is proportional to the depth of the intensity minimum (the depth is proportional to the concentration of NH_3).
5. The concentration of NH_3 can be measured by the amplitude of IMLS.

A good correlation (correlation coefficient (q) of 0.997) is obtained between the analytical values obtained by the NH_3 analyzer and those by the standard method (indophenol). The scattering deviation for coal-fired flue gas observed by the standard method ($s = 10.2$ ppm) is larger than the value obtained by the NH_3 analyzer ($s = 2.9$ ppm). The results obtained by using the NH_3 analyzer for coal-fired flue gas are shown in Figure 9.

Interference from SO_2 is shown in Figure 10. By subtraction of the interference signal, good correspondence has been obtained between the analytical values using the standard method and those obtained by the NH_3 analyzer.

To prove the reliability of the NH_3 analyzer, the device will be used in the demonstration test at Takehara No. 1 unit.

Future study of the NH_3 analyzer will concern:

1. The reliability of the measured value at low concentrations (such as 1 or 2 ppm).
2. Interference from SO_2 concentrations over 1,000 ppm compensated by employing an automatic calculation method.
3. Locating the NH_3 analyzer near the sample probe.

Section 4

DEVELOPMENT STATUS FOR FLUE GAS TREATMENT TECHNOLOGY AND LOW POLLUTION COAL-FIRED POWER STATIONS

COMMERCIALIZATION CLASSIFICATION AND DEVELOPMENT STAGE OF FLUE GAS TREATMENT TECHNOLOGY FOR COAL-FIRED BOILERS IN JAPAN.

EPDC has been executing R&D programs on flue gas treatment technology concerning emissions of dust, SO_x, and NO_x from coal-fired boilers. As a result there are numerous processes (specified in Table XIII) both under development and in the commercialization stage for application in Japan. The current status of development and/or commercialization is show in Table XIV.

MODEL OF A LOW POLLUTION COAL-FIRED POWER STATION

Numerous combinations of flue gas treatment systems are possible for the control of dust, SO_x, and NO_x from coal-fired power stations. Models of three combinations are shown in Figure 11, and are anticipated to meet target emission levels similar to that from oil-fired power stations (i.e., 60 ppm NO_x, 50 ppm SO_x, and 10 mg/Nm3 dust). Detailed specifications of these three models are presented in Table XV.

Among the three cases for SO_2 removal, the Dry $DeSO_x$ System (Case 3) is quite attractive to EPDC in terms of simplicity, economy, performance, and reliability. Thus, EPDC is highly interested in R&D for Dry $DeSO_x$ and it is a key project of Case 3. EPDC is planning to perform a demonstration test at the EPDC Matsushima Power Station (500 MW coal).

TREND OF EPDC'S R&D PROGRAM IN THE FUTURE

R&D on NO_x and SO_x is scheduled to be virtually complete by March 1981. However, dust emission control in Japan may be tightened, thus altering this schedule. Accordingly, dust collecting technologies such as bag houses and high performance ESPs with particle charging system are drawing attention.

EPDC has been promoting R&D of emission control technology for modern coal-fired power plants necessary to meet social needs. Establishing such technologies has received formost attention. In the near future, R&D will be directed to optimizing such processes for economy, reliability, and performance.

Section 5

DEMONSTRATION TEST AND COMMERCIALIZATION PLAN

After assessing the commercialization potential through pilot-scale development efforts, a demonstration test to identify commercial capabilities is needed. The Japanese government is sponsoring demonstration tests conducted by EPDC to achieve this end.

THE DEMONSTRATION DeNO$_x$ TEST OF TAKEHARA POWER STATION NO. 1 UNIT

EPDC's demonstration DeNO$_x$ plant sponsored by the Japanese government and initiated in 1978, is now under construction. Construction is 20% complete, and testing will be initiated in July 1981.

The concept of the Takehara No. 1 unit demonstration test is shown in Table XVI.

NH_3 Injection and Its Control

As the NH_3 injection system of the SCR demonstration equipment at Takehara No. 1 unit shows (Figure 12), NH_3 is mixed and diluted with air in the dilution mixing chamber and injected before the SCR. To keep the NH_3 concentration constant, the NH_3 injection system is designed as follows:

- Sufficient distance is maintained between the NH_3 injection point and the SCR unit to mix NH_3 adequately.
- Uniform velocity distribution is maintained by means of guide vane installation at duct bends, or installation of a gas adjustment grid upstream from the NH_3 injection point.
- A constant mixing ratio between NH_3 and dilution air is maintained or the diameter of the NH_3 injector nozzle can be optimized to keep this dilution concentration and dilution gas speed constant.

Control of the NH_3 feed rate is accomplished by injecting NH_3 at a constant ratio to the NO_x quantity in the flue gas. An input control defines the set value for NH_3/NO_x ratio. NO_x concerntration at reactor outlet is monitored with the NH_3 feed rate controlled by feed-back signals from the reactor outlet NO_x concentration. Figure 13 shows the control system for NH_3 injection.

TAKEHARA NO. 3 UNIT PLAN

Takehara No. 3 unit initiated construction in May 1980. Commercial operation will begin in March 1983. The Takehara No. 3 unit flow system (LDSS) is shown in Figure 14, with the layout in Figure 15. This facility will be largest coal-fired power plant in Japan. Levels of NO_x, SO_x, and dust anticipated to be emitted are shown on Table XXII and, as can be seen, are extremely low.

DRY $DeSO_x$ DEMONSTRATION TEST

EPDC is planning to install dry $DeSO_x$ demonstration test equipment at the Matsushima Power Station (700 MW coal) in April 1983. This demonstration test is funded by the Japanese Government. EPDC had designated Sumimoto Heavy Industries (SHI) to build the dry $DeSO_x$ system since EPDC has been executing an R&D project on dry $DeSO_x$ in cooperation with SHI.

Table XVIII shows the schedule and Table XIX shows outline of this demonstration.

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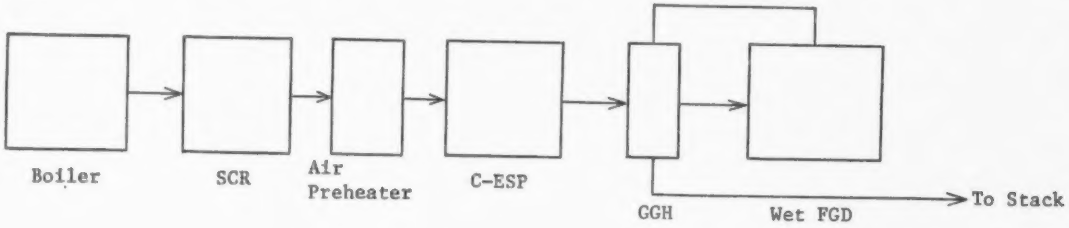
2. T. Kimura, Y. Nakabayashi, K. Mouri, Overall Flue Gas Treatment Technology from Coal Fired Power Plant in Japan, ICCR 5th Conference Dusseldorf, Sept. 1980.

Figure 1

High Dust Denitration System (HDDS) and Low Dust Denitration System (LDDS)

[1] HDDS


Boiler
SCR
Air Preheater
C-ESP
GGH
Wet FGD
To Stack

[2] LDDS

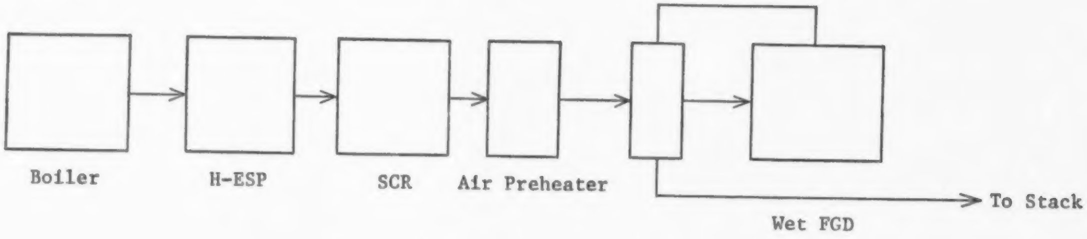

Boiler
H-ESP
SCR
Air Preheater
Wet FGD
To Stack

Figure 2

Problems Depend on Leakage NH_3 from SCR Process

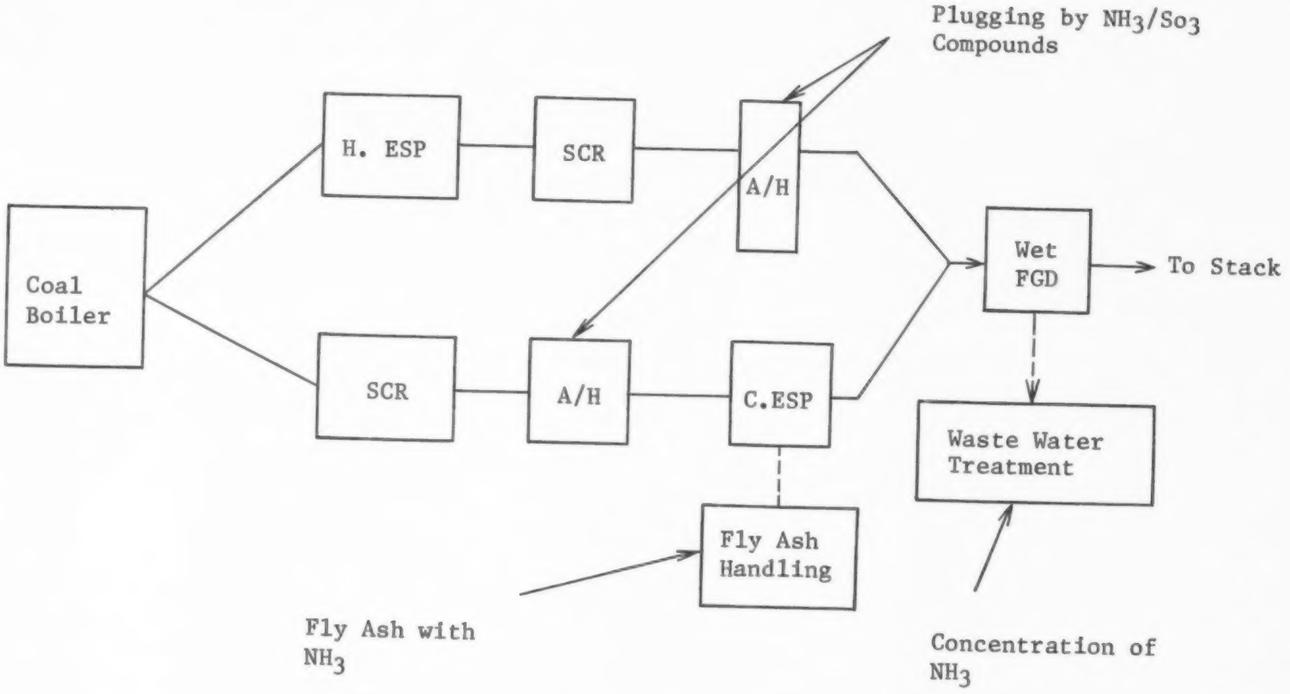

Plugging by NH_3/So_3 Compounds
Coal Boiler
H. ESP
SCR
A/H
SCR
A/H
C.ESP
Wet FGD
To Stack
Waste Water Treatment
Fly Ash Handling
Fly Ash with NH_3
Concentration of NH_3

Figure 3

Biological Nitrogen Removal Method in Waste Water from De-SOx Process

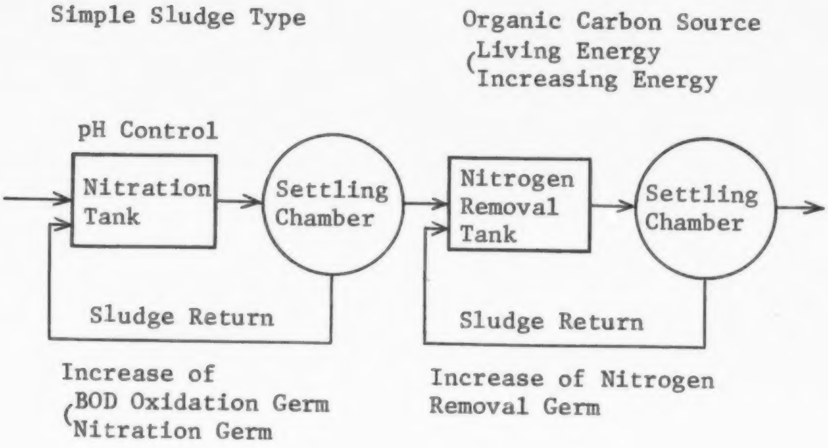

Simple Sludge Type
Organic Carbon Source
Living Energy
Increasing Energy
pH Control
Nitration Tank
Settling Chamber
Nitrogen Removal Tank
Settling Chamber
Sludge Return
Sludge Return
Increase of
BOD Oxidation Germ
Nitration Germ
Increase of Nitrogen Removal Germ

Figure 4

Experiment Flow Diagram of Air Preheater for Denitration Process

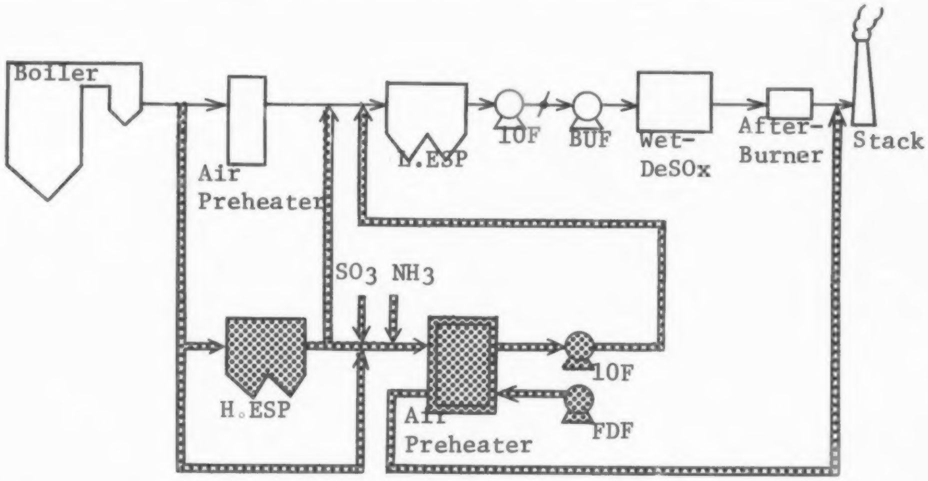
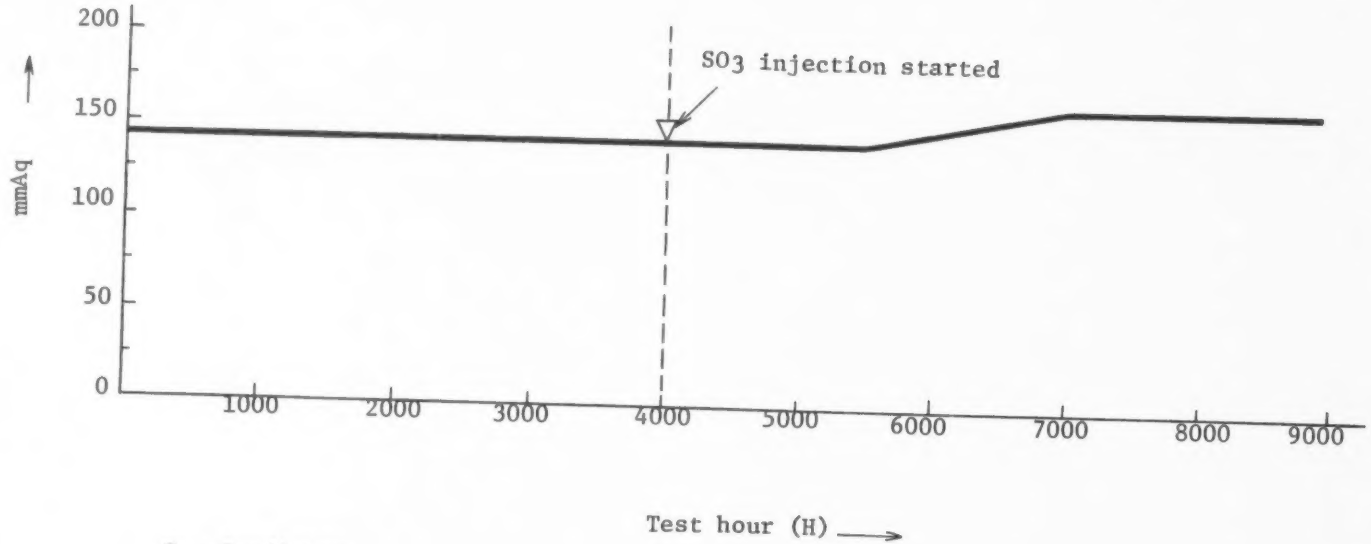

Boiler
Air Preheater
H.ESP
IOF
BUF
Wet-DeSOx
After-Burner
Stack
SO3 NH3
H.ESP
IOF
Air Preheater
FDF

Figure 5

Draft Loss Trend of Test Air Preheater and Conditions (Ljungstrom Test)



Gas Condition

SO_2	1200 - 1300 ppm	SO_2	1200 - 1300 ppm
SO_3	3 - 5 ppm	SO_3	20 ppm
Dust	0.1 - 0.2 g/Nm_3	Dust	0.1 - 0.2 g/Nm_3
NH_3	10 ppm	NH_3	10 ppm
Gas Temp of AH inlet	325-337°C	Gas Temp of AH inlet	325-337°C
Gas Temp of AH outlet	145-165°C	Gas Temp of AH outlet	145-165°C

Figure 6

One Sample of

SV vs Nox Curves

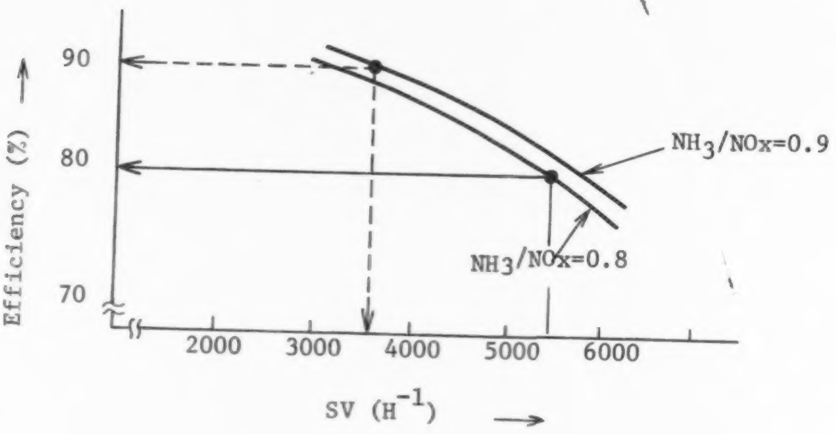


Figure 7

Absorption Spectrum of Ammonia

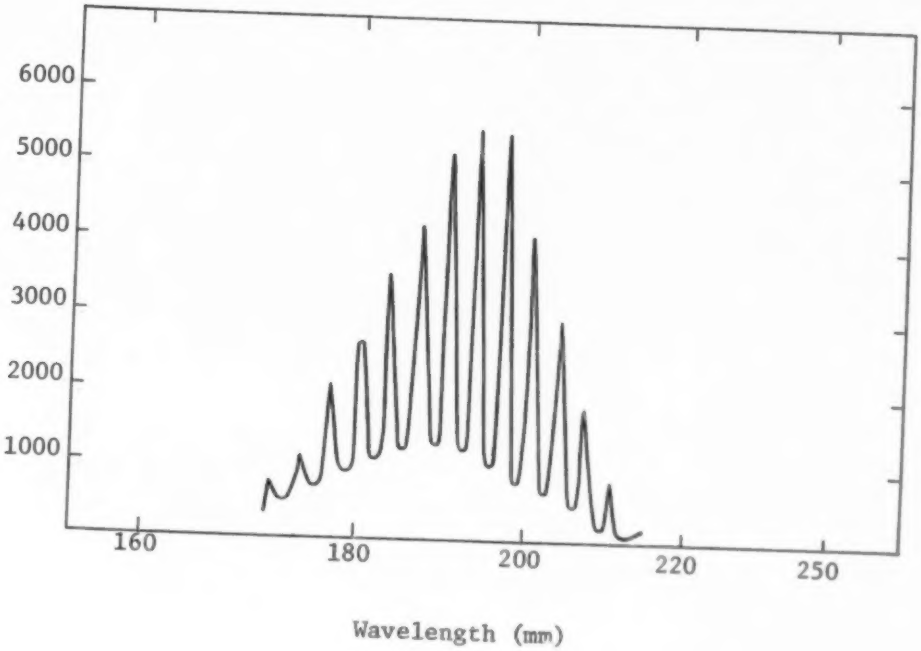


Figure 8

Principle

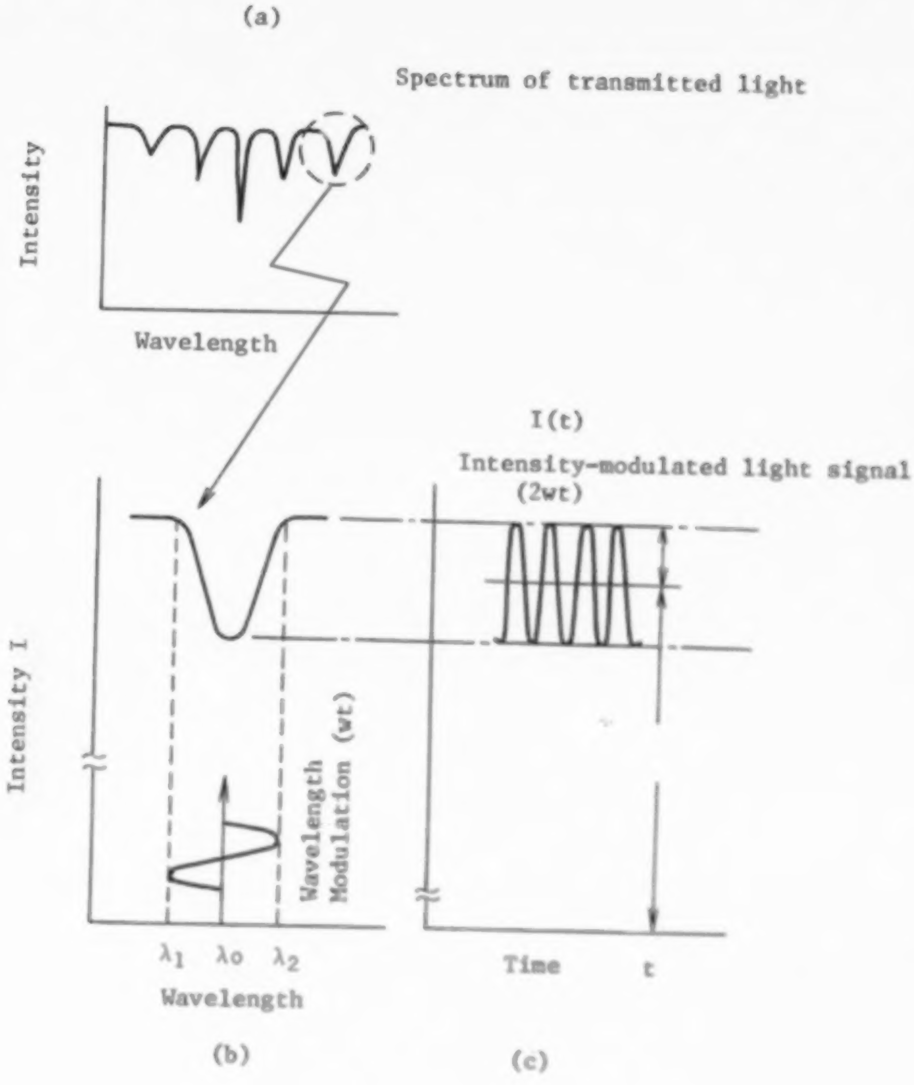

(a)
Spectrum of transmitted light
Intensity
Wavelength
I(t)
Intensity-modulated light signal
(2wt)
Intensity I
Wavelength
Modulation (wt)
λ1
λo
λ2
Wavelength
Time
t
(b)
(c)

Figure 9

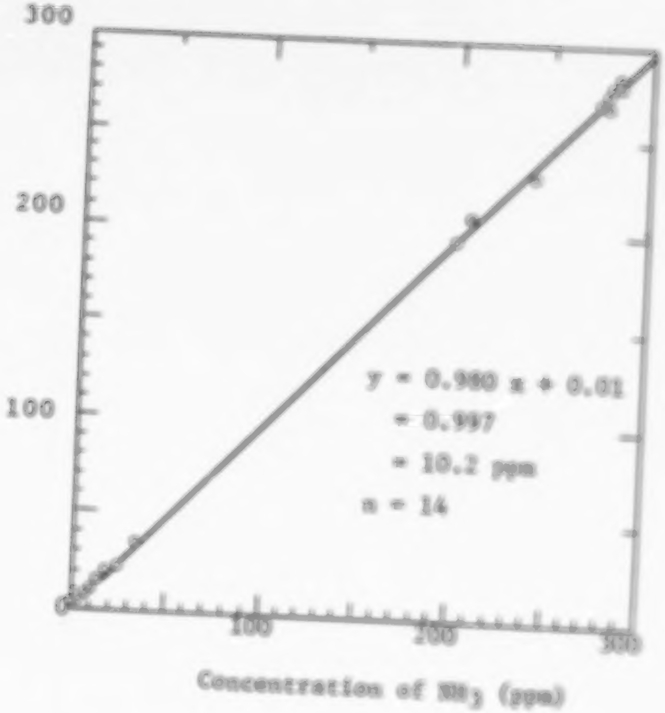


Figure 10

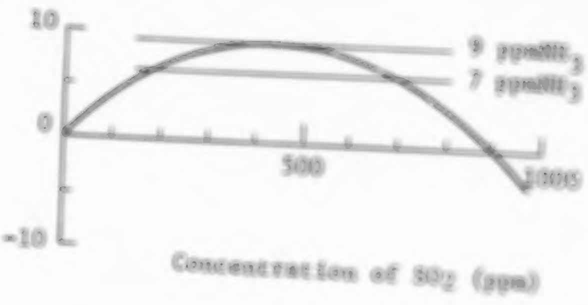


Figure 11

Model Anti-pollution Coal-fired Thermal Power Plant

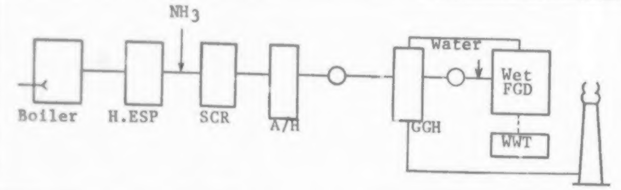
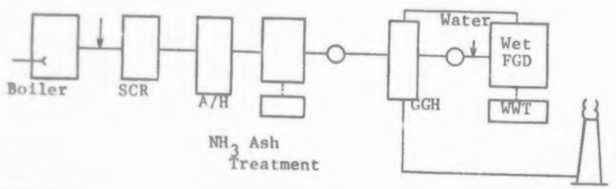
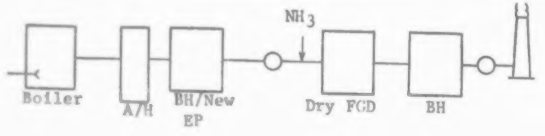
Case	System	Power Plant Flow	Con-centration	Remarks
1	Low Dust Denitration System	 NH3 Water Wet FGD Boiler H.ESP SCR A/H GGH WWT	-	H.ESP: Hot-side Electrostatic Precipitator SCR: Selective Catalyst Reduction Equipment
2	High Dust Denitration System	 Water Wet FGD Boiler SCR A/H GGH WWT NH3 Ash Treatment	-	A/H: Air Preheater GGH: Gas-Gas Heat Exchanger FGD: Flue Gas Desulfuriza-tion
3	Dry FGD System	 NH3 Boiler A/H BH/New EP Dry FGD BH	-	W.WT: Waste Water Treatment Equipment BH: Bag House O: Fan

Figure 12

NH_3 Injection Flow

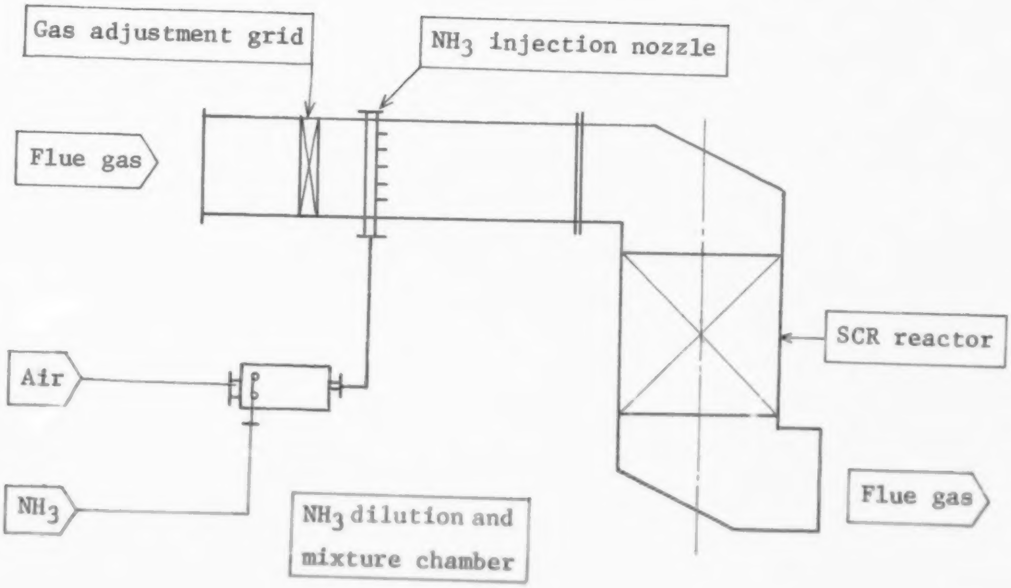

Gas adjustment grid
NH3 injection nozzle
Flue gas
SCR reactor
Air
NH3
NH3 dilution and mixture chamber
Flue gas

Figure 13

Fundamental Control System for NH_3 Injection

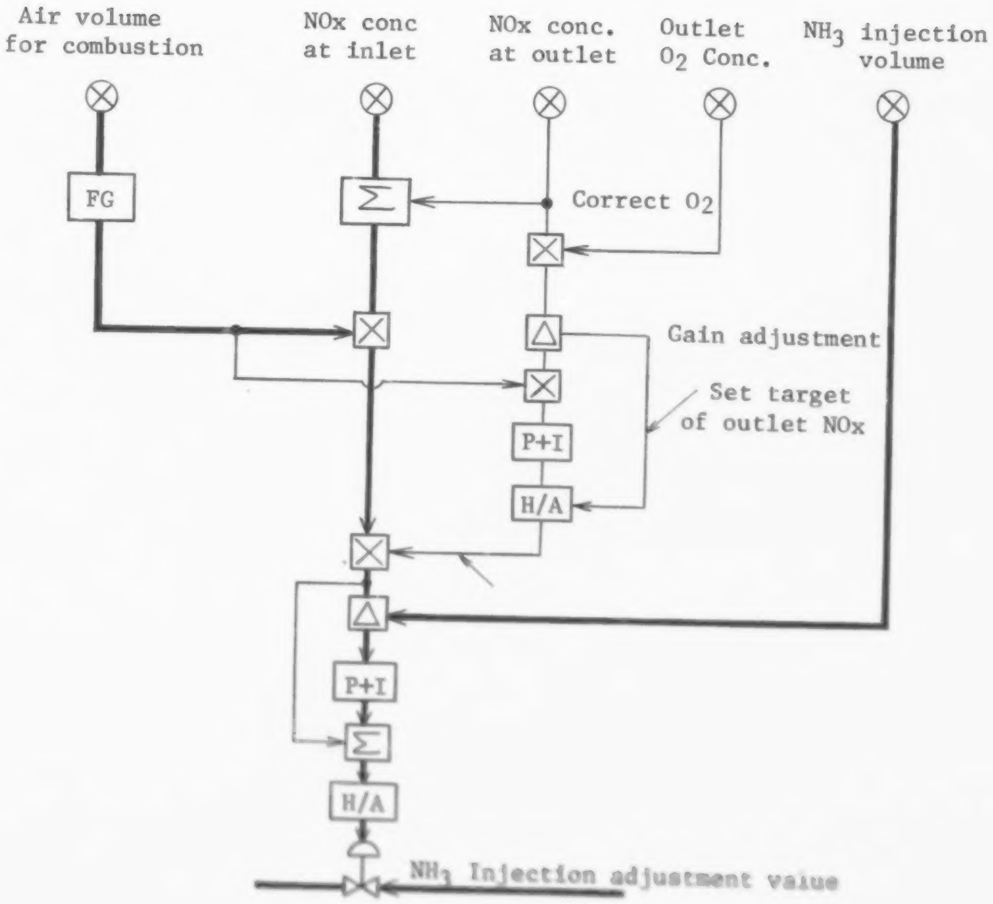

Air volume for combustion
NOx conc at inlet
NOx conc. at outlet
Outlet O_2 Conc.
NH_3 injection volume
FG
Σ
Correct O_2
Gain adjustment
Set target of outlet NOx
P+I
H/A
P+I
Σ
H/A
NH_3 Injection adjustment value

Figure 14

Overall Flue Gas Treatment System

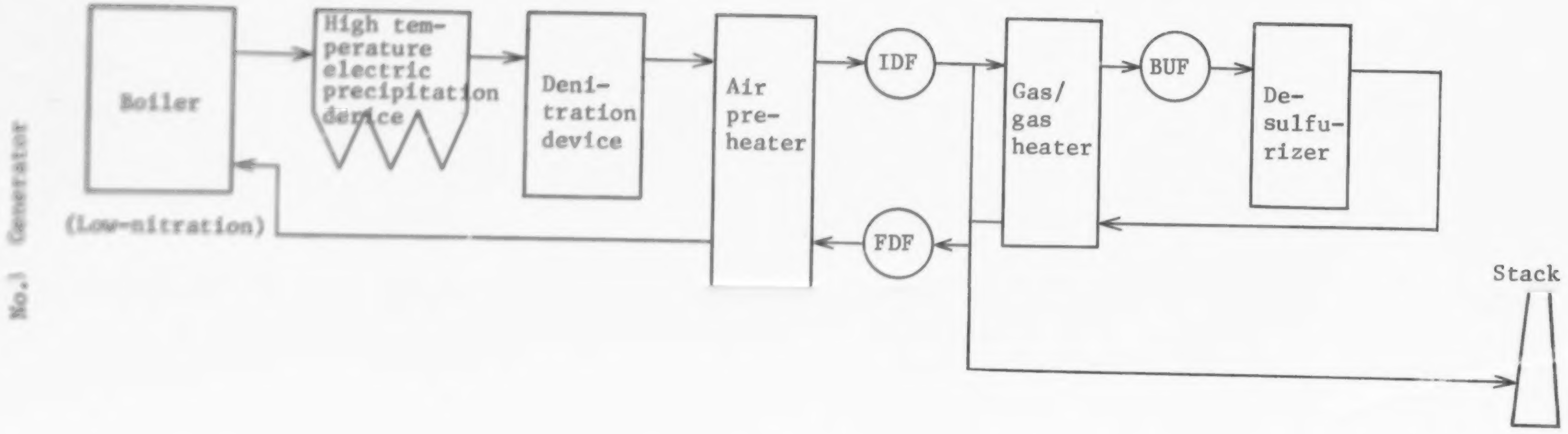

No.1 Generator
Boiler
(Low-nitration)
High temperature electric precipitation device
Denitration device
Air preheater
IDF
FDF
Gas/gas heater
BUF
Desulfurizer
Stack

Table I DeNOx Installation in the Power Utilities of Japan

Power Company	Process	Manufacture	Power Plant	Unit	Out Put (MW)	Fuel	Start-up	Gas Volume (Nm3/H)	Capacity (%)	Efficiency (%)	Remarks
Hokkaido EPCO	SCR	BHK	Tomato-Azumi P/S	1	350	Coal	Sep. 1980	280,000	25	90	Test Equipment
Tohoku EPCO	SCR	IHI	Higashi-Niigata P/S	2	600	H.O, Crude, LNG	Aug. 1981	1,660,000	100	—	
Tokyo EPCO	SCR	MHI	Yokosuka	4	350	H.O	Feb. 1978				
Chubu EPCO	SCR	BHK	Chita	5	700	LNG	Mar. 1978	1,910,000	100	80	
	SCR	BHK	Chita	6	700	LNG	Mar. 1978	1,910,000	100	80	
	SCR	MHI	Chita	4	700	H.O	Nov. 1979	1,960,000	100	80	
	SCR	MHI	Atsumi	3	700	H.O	Dec. 1980	1,900,000	100	80	
	SCR	MHI	Atsumi	4	700	H.O	Feb. 1981	1,900,000	100	80	
	SCR	MHI	Shin-Nagoya	3	220	H.O	Jul. 1980	650,000	100	80	
	SCR	IHI	Shin-Nagoya	6	500	H.O, Crude, etc.	Jun. 1980	1,316,000	100	—	
Hokuriku EPCO	SCR	IHI	Toyama-Shinko	2	500	H.O, Crude	Nov. 1981	1,370,000	100	—	
Kansai EPCO	SCR	BHK	Kainan		450	H.O	Jun. 1977	300,000	25	80	
	Compact SCR	BHK	Amagasaki-Higashi	1	156	H.O	Jul. 1978	466,000	100	30	
	Compact SCR	MHI	Osaka	1	156	H.O	Jun. 1978	490,000	100	30	
	SCR	MHI	Osaka	3	156	H.O	Jul. 1980	552,000	100	80	
	SCR	MHI	Osaka	4	156	H.O	Dec. 1979	552,000	100	80	
	SCR	MHI	Sakaiko	1	250	H.O	Jul. 1980	800,000	100	80	
	SCR	MHI	Sakaiko	6	250	H.O	Dec. 1979	800,000	100	80	
	SCR	MHI	Osaka	2	156	H.O	Jul. 1981	530,000	100	80	
	SCR	MHI	Tanagawa	3	156	H.O	Aug. 1981	520,000	100	80	
	SCR	MHI	Tanagawa	4	156	H.O	Mar. 1981	520,000	100	80	
	SCR	MHI	Sakaiko	2	250	H.O	Sep. 1981	800,000	100	80	
	SCR	MHI	Sakaiko	4	250	H.O	Feb. 1981	800,000	100	80	
	SCR	MHI	Sakaiko	7	250	H.O	Sep. 1981	800,000	100	80	
	SCR	IHI	Himeji	5	156	H.O	Jun. 1978	490,000	100	—	
Chugoku EPCO	SCR	MHI	Iwakuni	2	350	H.O	Dec. 1980	980,000	100	80	
	SCR	MHI	Iwakuni	3	500	H.O	Apr. 1981	1,400,000	100	80	
	SCR	MHI	Shimonoseki	1	175	Coal	Apr. 1979	550,000	100	60	(80% in near future)
	SCR	IHI	Kudamatsu	2	375	NGL, H.O, Crude	Apr. 1979	1,050,000	100	—	
	SCR	IHI	Kudamatsu	3	700	NGL, H.O, Crude	Sep. 1979	1,900,000	100	—	
Kyushu EPCO	SCR	MHI	Shinkokura	3	600	LNG	Jun. 1978	1,690,000	100	>75	
	SCR	MHI	Shinkokura	4	600	LNG	Jun 1979	1,690,000	100	>75	
EPDC	SCR	BHK, KHI	Takehara	1	250	Coal	Jul. 1981	400,000x2	100	80	
	SCR	Not decided	Takehara	3	700	Coal	Mar. 1983	2,414,000	100	80	
Joban Kyodo EPCo	SCR	IHI	Nakoso	9	600	Coal, H.O	Apr. 1983	1,700,000	100	—	
	SCR	MHI	Nakoso	8	700	Coal, H.O	Dec. 1982	1,700,000	100	>50	(80% in near future)
		BHK	—	—	Total	H.O	July, 1978~ July, 1980	Total 2,586,600	—	—	
		MHI	—	—	Total 306	Coal	June, 1982~ July, 1982	Total 1,166,000	—	—	
		IHI	—	—	Total 2387	H.O, Crude	April, 1978~ May, 1982	Total 6,848,000	—	—	

Table II Comparison of DeNOx Processes

Process	Economy	Performance		Secondary Pollution	Reliability			Development
		NOx	SOx		Operation	Maintenance	Danger	
SCR	3	>80%	>90%	None	Easy	Easy	None	1∿2 years
SNR	1	>30%	>90%	None	Easy	Difficult	Hydrogen use	1∿2 years
SCR+SNR	2	>60%	>90%	None	Easy	Difficult	None	1∿2 years
Dry DeSOx/ DeNOx (CuO Type)	4	>80%	>80%	None	Difficult	Difficult	Hydrogen use	3∿4 years
Wet DeSOx/ DeNOx	5	>80%	>90%	By Product as NO_3	Difficult	Difficult	None	1∿2 years

Table III Feature of Catalyst for LNG, Oil and Coal

AV: Area Velocity

Fuel	Countermeasure		Shape	Cost to 1 unit
	Dust	Sox		
LNG	None	None	Pellet	1
Oil	None	None or Need	High AV Honeycomb, pipe board etc.	2
Coal	Need	Need	Low AV Honeycomb, pipe board etc.	3

Table IV SCR Pilot Tests for Coal Fired Boiler

Company	Joint research	Power plant	Capacity (Nm^3/H)	Shape of catalyst	HDSS or LDSS	Schedule 1976	(Fiscal Year) 1977	1978	1979	1980	1981
EPDC	IHI	Isogo	1000 x '2	Honeycomb	HDSS			━	━	━	━
			1000 x 2		LDSS			━	━	━	━
	H Z	Isogo	230	Honeycomb	HDSS	━	━	━	━	━	━
			140 x 2		LDSS	━	━	━	━	━	━
	MHI	Takasago	200 x 2	Honeycomb	HDSS	━	━	━	━	━	
			600 x 2		LDSS	━	━	━	━	━	
	KHI	Takehara	250 x 2	Pipe	HDSS	━	━	━	━	━	━
			4200		LDSS			━	━		
	BHK	Takehara	2300	Board	LDSS			━	━		
			1330		HDSS					━	
Tokyo EPCO	MHI	Nakoso (1)	4000	Honeycomb	HDSS				━	━	
			4000		LDSS				━	━	
Hokkaido EPCO	BHK	Ebetsu	2000	Ring (Moving Bed)	LDSS	━					

(1) Fuel of Nakoso is 70% oil and 30% coal.

Table V Comparative Items between HDSS and LDSS

Equipment	Items
(1) Overall System	Plant Efficiency, Layout, Duct Work Countermeasure for GRF erosion Countermeasure for A/H Plugging and Draft Loss, Treatment of Fly Ash with NH_3, Behavior of SO_3
(2) Precipitator	Treated Gas Volume, Efficiency, Size Heat Expansion, Quality and Properties of Fly Ash
(3) SCR	Erosion of Catalyst, Shape of Catalyst Catalyst Volume, Catalyst Life, Production Method of Catalyst, Gas Velocity Reactor Structure, Draft Loss, Leakage NH_3
(4) Wet DeSOx	Gas Heat Exchanger Waste Water Treatment

Table VI Comparison between HDSS and LDSS (Overall System)

Items	HDSS	LDSS
° Plant Efficiency	Base	Decline
° Layout	Base	Larger
° Countermeasure for Erosion	Use Multi Cyclone	Not necessary
° Countermeasure for A/H Plugging	① Reduction of leakage NH_3	① ditto ② Low SO_2/SO_3 conversion catalyst ③ Improved A/H for SCR
° Draft Loss	Base	Increase
° Treatment of Fly Ash with NH_3	Contained NH_3 into Fly Ash Need NH_3 removal treatment under contain circumstances.	No trouble

Table VII Comparison between HDSS and LDSS (II) (Precipitator)

Item	HDSS	LDSS
• Gas Volume	Base (130 - 150°C)	Approx. 1.8 times (370 - 380°C)
• Efficiency	Base	Applicable to various kinds of coal.
• Size	Base	Generally bigger
• Heat Expansion	Small	Large
• Quality of Fly Ash	Contain NH_3 but No trouble	No problem
• Treatment of Fly Ash	Simple device [illegible] low temperature ash	Need insulator, burn protections and ash cooler for high temperature ash.

Table VIII Comparison between HDSS and LDSS (III)
(SCR)

Item	HDSS	LDSS
• Influence by Dust	Need anti-erosion for catalyst	No problem
• Shape/peculiarity of Catalyst	Parallel passage type Such as honeycomb, pipe board etc. and low erosion material	Same
• Catalyst Quantity	Base	Nearly Same
• Catalyst Life	2 - 3 years (Presumption)	3 - 4 years (Presumption)
• Product Method of Catalyst	Base	Same
• Gas Velocity	Base	Same
• Draft Loss	Base	Same
• Leakage NH_3	Base	Same

Table IX Comparison between HDSS and LDSS (IV)
(DeSOx)

Item	HDSS	LDSS
• DeSOx	No problem	No problem
• GGH Plugging/ Corrosion	Less countermeasure for plugging/corrosion to LDSS as NH_3 and SO_3 is precipitated by ESP	Need the counter-measure such as strong soot blow, element material etc.
• Waste Water Treatment	Need De-N device to blow dow	De-N device is 2 or 3 times to HDSS

Table 8 De-N Development for DeSOx Waster Water

Company	Joint Researcher	Process	Capacity	Test Location	Schedule (Fiscal year)			
					1977	1978	1979	1980
EPDC	Hitachi Zosen (HZ)	Biological	Labo. test Max 20l/D	HZ's Labo	▬			
			Field test Max 5 m^3/D	Takehara P/S		▬		
			Labo. test 3l/D	HZ's Labo (1)			▬	
	Mitsui-Miike Machinery (MMM)	Biological	Labo. test	MMM's Labo	▬			
			Field test	Ditto		▬		
			Labo. test	Ditto (1)			▬	
	Ebara Infilco	Biological	Labo. test	EI's Labo	▬			
			Field test	Ditto		▬		
			Labo. test	Ditto (1)			▬	
	Kurita (K)	Biological	Labo. test	K's Labo	▬			
	Organo	Biological	Labo. test	O's Labo	▬			

Note: (1) De-N and De-COD simultaneously.

Table XI Outline of Air Preheater Test

Company	Joint Researcher (Type of A/H)	Test Location	HDSS or LDSS	Capacity (Nm^3/H)	Schedule (Fiscal year)				
					1976	1977	1978	1979	1980
EPDC	Gadelius (Ljungstrom)	Takasago P/S	HDSS	10,000	▬				
			LDSS	5,000		▬			
			LDSS	5,000			▬		
			LDSS	8,000				▬	▬
	Hitachi Zosen (HZ) (Rothemühle)	Takehara P/S	LDSS	2,000			▬	▬	
		Isogo P/S	LDSS	-					====
	IHI (Rothemühle)	Isogo P/S	HDSS	2,000			▬	▬	▬
			LDSS	2,000			▬	▬	▬
Tokyo EPCO	MHI	Nakoso P/S	HDSS	4,000				▬	▬
			LDSS	4,000				▬	▬

Table XII Identification of A/H Plugging

Condition	High dust No NH_3 injection	High dust NH_3 injection	Low dust NH_3 injection	Low dust NH_3 injection
Plugging	None	None	None	Yes
Test Condition	15g/Nm^3	15g/Nm^3 10 ppm	0.1g/Nm^3	0.1g/Nm^3 10 ppm

Table XIII Flue Gas Treatment Techniques

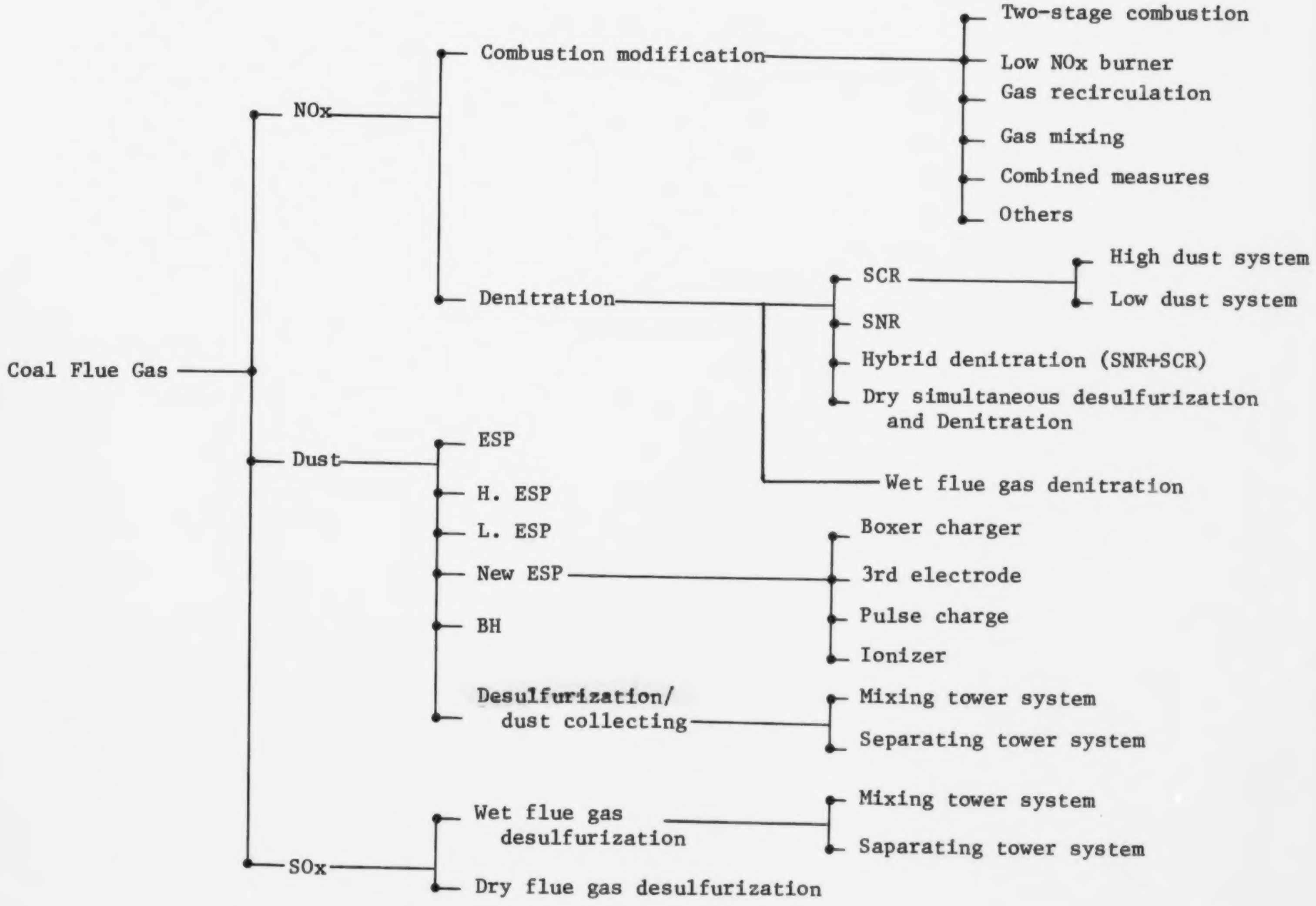

Coal Flue Gas
NOx
Combustion modification
Two-stage combustion
Low NOx burner
Gas recirculation
Gas mixing
Combined measures
Others
Denitration
SCR
High dust system
Low dust system
SNR
Hybrid denitration (SNR+SCR)
Dry simultaneous desulfurization and Denitration
Wet flue gas denitration
Dust
ESP
H. ESP
L. ESP
New ESP
Boxer charger
3rd electrode
Pulse charge
Ionizer
BH
Desulfurization/ dust collecting
Mixing tower system
Separating tower system
SOx
Wet flue gas desulfurization
Mixing tower system
Saparating tower system
Dry flue gas desulfurization

Table XIV Present Status of Research and Development (1/2)

	Boiler		H.EP	S C R			A/H		Remarks
	Combustion modification	S N R		High dust	Low dust	Regeneration	High dust	Low dust	
(1) Steps of research and development	Under R & D	Ditto.	in practical use	under R & D	practicable	R & D	practicable	practicable	SNR: Selective Non-Catalytic Reduction SCR: Selective Catalytic Reduction
(2) Co-researchers	Babcock Hitach, IHI, MHI, KHI	MHI, IHI	SHI,(Gadelius Hitachi, MHI)	HZ, IHI, MHI, KHI, Hitachi	Ditto	KHI	Gadelius IHI	Gadelius, IHI, HZ	
(3) Development targets	100ppm and below (6%O_2) 5% and below in unburned carbon, with CO and HCN showing no increase.	40% and above 100ppm and below	100mg/Nm^3 and below	80% and above 5ppm and below 0.5 - 1.0% and below (performance after the lapse of one year)	Ditto	Ditto	To operate stably for one year or more.	Ditto.	
(4) Results so far achieved	Guidelines for NOx reduction measures have been set up and improvements have been made through testing by type of coals.	° NOx is not affected by fly ash. ° Furnace temperatures have been actually measured. (Isogo P/S)	° Specifications necessary for design of H.ESP have been obtained.	Development targets have been cleared.	Ditto	° Fundamental techniques for regeneration have been established. ° Secular changes in performance are being checked after completion of flue gas regeneration testing.	° Tendency toward plugging has been confirmed.	° Soot blowing has been intensified. And element has been improved.	
(5) Subjects in the future	To develop a boiler of 100ppm and below ° To develop an ultra-low NOx burner ° To make a survey of optimum condition	° To study a simple denitration means, ° To study cooling systems and ° To develop an NH_3 ash disposal process.	° To investigate the effects of Na and Ca.	° To develop ammonia ash disposal process	(Optimum design)	° Secular change after regeneration	° Concrete ideas to improve commercialized equipment	To make a survey of the optimum design	
(6) Time of putting into practical use	200ppm can be guaranteed at present.	° After developing the NH_3 ash disposal process	Under construction in Matsushima Power Station	After developing the ammonia ash disposal process	Demonstration test plant is under construction in No.1 unit, Takehara Power Station.	Not fixed yet	Practicable	Demonstration test plant is under construction in No.1 unit, Takehara Power Station.	

Present Status of Research and Development (2/2)

	Dust Collector		G G H	Desulfurization		Wet EP	A/B	B H	Remarks
	Boxer Charger	B.H.		Wet	Dry				
(1) Steps of research and development	Under R & D	Ditto.	In practical use	Already put into practical use	Under R & D	Ditto	Already put into practical use	Under R & D	
(2) Co-researchers	IHI(Toshiba) Hitachi	SHI(IHI)	Gade, IHI	-	SHI,(IHI)	IHI(SHI)	-	SHI	
(3) Development targets	10mg/Nm^3 and below	10mg/Nm^3 and below	To operate stably for one year or more.	(95% and above)	95% and above (ηnox 40% and above)	10mg/Nm^3 and below	-	10mg/Nm^3 and below	
(4) Results so far achieved	Basic data has been gathered	Laboratory tests have been completed.	Design conditions with and without NH$_3$ have been confirmed.	-	ηSOx and SOx performance has been already confirmed.	-	-	Performance confirmation	
(5) Subjects in the future	Testing with pilot plant for confirmation of practical use	To make sure of filter cloth life and performance and to optimize design conditions	Optimum design (measures for clogging with NH$_3$)	Performance of collecting submicron particules, SO$_3$, etc.		Performance confirmation		Optimum design	
(6) Time of putting into practical use	Not fixed yet	Not fixed yet	Under construction in Matsushima Power Station (Case without leakage of ammonia) No.3 unit under construction in Takehara Power Station (Case with leakage of ammonia)	Already put into practical use	Demonstration test plant is expected to be provided in Matsushima Power Station. (Entrusted by government)	Not fixed yet	-	Demonstration test plant is expected to be provided in Matsushima Power Station. (In a combination with dry FGD)	

Table XV Features of Anti-Pollution Coal-Fired Power Plant by System

Case	System	Flue Gas Emission Level	Prospects of Practical Use	Denitrification	Countermeasure for A/H	Ammonia Ash Treatment	Layout	Combustion Modification
1.	Low dust denitration	NOx 60 ppm and below SOx 100 ppm and below Dust 30 mg/Nm^3	Practicable (700 MW under construction in Takehara P/S)	Required	Required	Not required	Base	Combustion modified to 300 ppm and below in NOx. Already put into practical use.
2.	High dust denitration	Ditto	Within several years. (Ammonia ash treatment techniques have not been established yet.)	Required	Not especially required.	Required	Better	Ditto
3.	Dry Desulfurization	NOx 60ppm and below SOx 100ppm and below Dust 1 mg/Nm^3	Within several years	Not required	Not required	Required	Best	NOx must be reduced to 100 ppm. A modification means is being developed.
	Existing							

Table XVI Outline of SCR Demonstration Plant at Takehara No. 1 Unit

Item	Manufacture	Type	Capacity	Efficiency	Remarks
(1) Existing Plant					
(a) Boiler	BHK	Reheat Type Radiant Boiler	810 T/H	-	Fuel: coal
(b) Plant output	-	-	250 MW	-	
(2) Demonstration Plant					
(a) SCR	BHK KHI	Board Cat. Pipe Cat.	400,000 Nm^3/H 400,000 Nm^3/H	>80% >80%	
(b) Air Preheater	Gadilius	Long element Ljumgstrom	800,000 Nm^3/H	-	Now Air Preheater
(c) De-N	HZ	Biological Process	1,000 m^3/Day	<10ppm as total N	

Table XVII Flue Gas Emissions from Takehara No. 3 Unit

ITEM	Unit	Numerical Value
Gas volume (Wet) (Dry)	Nm^3/H	2,414,000 2,169,000
Discharge temperature	°C	100
Stack height	m	200
SOx concentration	ppm	100
NOx concentration	ppm	60(O_2=6%)
Dust	g/Nm^3	0.03

Table XVIII The Demonstration Schedule

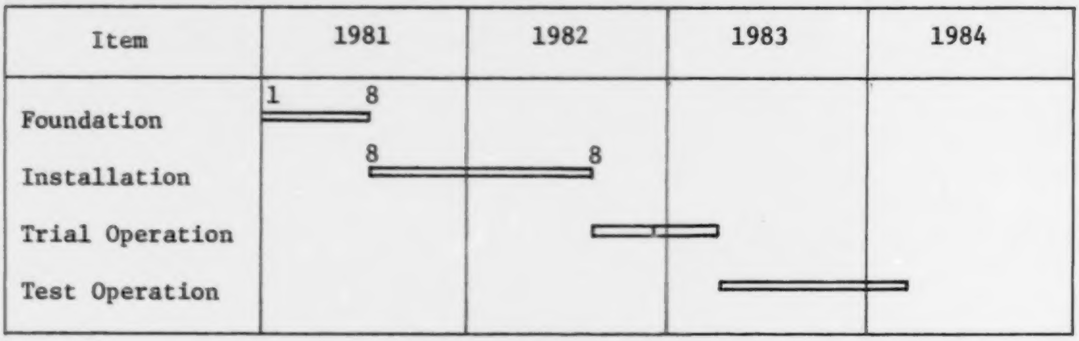


Table XIV

Outline of Dry DeSOx Demonstration Test Equipment

Item	Contents
° Process	Dry Type Activated Carbon Absorption
° Gas Volume	300,000 Nm^3/H
° Gas Temp.	135°C
° SO_2 (Inlet)	1,000 ppm
° SO_2 (Outlet)	50 ppm and below
° NO_x (Inlet)	300 ppm
° NO_x (Outlet)	225 ppm and below
° Dust (Inlet)	300 mg/Nm^3
° Dust (Outlet)	10 mg/Nm^3
° By product	Element Sulfur (purity 99.9% <)

STATUS OF SCR RETROFIT AT SOUTHERN CALIFORNIA EDISON HUNTINGTON BEACH GENERATING STATION UNIT 2

By:

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ABSTRACT

Utilities in the Southern California South Coast Air Basin are subject to a regulation (Rule 1135.1) requiring 90 percent NOx reduction. Rule 1135.1 is comprised of four basic compliance options of which the first two options require two stages with an intermediate milestone reduction and a demonstration unit of a 90 percent NOx reduction system.

This paper describes the Selective Catalytic Reduction (SCR) 107.5 MW Demonstration Facility SCE plans to install on one-half of Southern California Edison's Huntington Beach Unit 2, 215 MW boiler. The physical size, operation and maintenance, and controls for achieving 90 percent NOx reduction through normal load variations as well as the status of the project are discussed.

The system retrofit requirements are discussed with specific reference to the differences between the demonstration unit and other larger units and the site constraints for retrofit on the larger units. The operational and maintenance requirements for a systemwide retrofit and potential problem areas will also be reviewed.

The paper will present cost estimates for the Huntington Beach demonstration facility as well as SCE's projection of cost for adding SCR on the majority of its oil-fired units in the South Coast Air Basin. These costs will include capital as well as O&M. All costs will be presented in 1981 dollars.

I. INTRODUCTION

Utilities in the Southern California South Coast Air Basin are subject to a regulation (Rule 1135.1) requiring 90% NOx reduction. Over the past two years, many workshops and hearings have been held regarding the requirements of the rule. The latest version, adopted earlier this year, is comprised of four basic compliance options of which the first two options require two stages with an intermediate milestone reduction and a demonstration unit of a 90% NOx reduction system.

The demonstration unit requires the 90% NOx reduction on a unit of at least 100 megawatts in the Los Angeles basin by January 1, 1982. Depending on the options selected, a 90% NOx reduction basin wide must be achieved by 1988 or 1990. Considerable work is underway by the Southern California Edison Company to comply with the demonstration unit and much effort has gone into determining the impact of complying with the balance of the rule. This report describes the work performed to date.

II. HUNTINGTON BEACH SCR DEMONSTRATION PROJECT

FACILITY DESCRIPTION

The Huntington Beach Demonstration Facility employs a Selective Catalytic Reduction (SCR) process, in which nitrogen oxides in the boiler flue gas are reduced, using ammonia as a reducing agent. This facility will be capable of treating one half of the flue gas generated by Southern California Edison's Huntington Beach Unit 2, which is rated at 215 MW.

The SCR NOx removal process is depicted in Figure 1, and consists of the following five principle systems:

- A reactor system consisting of a reactor to contain and support the catalyst modules, a ducting/damper system to divert flue gas through or past the reactor, and a reactor booster fan with bypass duct to draw the flue gas through the reactor.

- An ammonia supply system consisting of a liquid ammonia storage tank, ammonia vaporizer and associated piping and controls to forward the ammonia vapor to the ammonia dilution skid.

- An ammonia dilution and injection system consisting of a blower for diluting the ammonia with combustion air from the air preheater, and an ammonia flue gas mixer to inject the diluted ammonia vapor uniformly into the flue gas stream.

- A control system consisting of ammonia injection controls, reactor booster fan control systems, a system to protect the boiler against negative pressure excursions and an emission monitoring system.

- An electrical system utilizing the existing Unit 5 gas turbine auxiliary transformer which includes a 4160 V switchgear, 480 V switchgear, lighting facilities, communication system, grounding system, DC power system and all associated raceways and cable.

EQUIPMENT

Reactor

The reactor is designed for vertical flue gas flow and has a fixed catalyst bed. The catalyst bed is built from individual catalyst modules which can easily be removed from the reactor through a large access door utilizing a permanently installed hoist system.

Reactor Booster Fan

The reactor booster fan is a centrifugal fan with air foil blading and is located downstream of the air preheater. The fan is directly coupled to a one speed electric motor and equipped with inlet vanes for flow control. A fan bypass duct and damper are provided to allow isolation of the fan from the existing boiler draft system.

Ammonia Tank

The ammonia tank is a horizontal vessel designed for storage of liquid anhydrous ammonia. The tank is sized to satisfy maximum ammonia demand for approximately 14 days of continuous operation. The tank is equipped with an external electric vaporizer to insure adequate ammonia vaporization at high demand/low ambient temperature conditions.

Equipment Data

Pertinent data for the above Apparatus as well as Huntington Beach Unit 2 are presented in Tables I and II.

ARRANGEMENT

Refer to Figures 2 and 3 for plan and elevation views of the NOx removal system. The arrangement takes advantage of an existing duct elbow between the economizer outlet and inlet to the hot side of the vertical shaft air preheater. This duct will be replaced with new ducting including branches for reactor inlet and outlet, and a reactor bypass. The bypass will allow boiler operation during reactor inspection and service.

A portion of the stack breaching will be removed to accommodate the reactor booster fan and fan bypass duct. The fan can discharge either to the existing stack or, through damper manipulation, to its own new stub stack.

The latter discharge mode will allow visual monitoring of the treated flue gas opacity.

The reactor is located in a vertical position directly south of the unit and does allow for system expansion to treat all flue gas flow from Unit 2.

CONTROLS AND INSTRUMENTATION

Ammonia Injection

A control system will be installed at Huntington Beach to inject ammonia according to unit load and flue gas NOx content at a rate to maintain a preset 90% removal ratio and a NH_3 slip within 0 to 10 ppm. To set the NOx removal at 90%, NOx will be monitored before and after the reactor for calculation of the removal ratio. The differential signal between the calculated ratio and the setpoint will be fed back to the NH_3 injection controls for correction of the injection rate.

Reactor Flue Gas Flow

A reactor booster fan is used to control the flue gas flow through the reactor and balance the flow between the east and west air preheaters. Air flow is controlled by inlet vanes which respond to boiler load and reactor differential pressure. The latter signal represents the actual flow through the reactor.

Boiler Protection

The addition of the reactor booster fan has increased the occurrence of boiler negative pressure excursions with an associated increased risk of boiler implosion. Hence, a protective control system will be added to mitigate this increased risk. This control system will take the reactor booster fan out of the flue gas loop in response to boiler trips, low furnace pressure, FD fan trips and low reactor outlet pressure. The booster fan draft is eliminated by the opening of fast acting dampers in the booster fan by-pass duct and the fast closing of the fan isolation damper.

Emission Instrumentation

Emission of NOx, NH_3, SO_2 as well as O_2 levels will be monitored downstream of the air preheater. In addition, opacity of the flue gas subjected to the SCR process will be visually monitored at the new stub stack.

TEST PROGRAM

The test program to be conducted at the Huntington Beach NOx removal facility will be designed primarily to determine the following process and operating parameters:

- NOx removal efficiency as a function of operating hours, ammonia injection rate, and NH_3 slip. A removal rate of 90% for 7,000 hours with an NH_3 slip of less than 10 ppm will be attempted.

- Assessment of catalyst operating life. This is a most important parameter with a dominating impact on system operating cost.

- Reactor and air preheater plugging as a result of deposits such as ammonia bisulfate.

- Operating factors such as reactor draft loss and ammonia consumption.

- Evaluation of ammonia injection controls, draft system controls, boiler protection controls and emission monitors and analyzers.

PROJECT STATUS

As indicated by the basic project schedule shown in Figure 4, all engineering has been completed and the construction contract awarded. All major equipment has been procured and will be delivered to the site during the last quarter of 1980. One exception is the reactor itself, which is being fabricated in Japan at this time, and is scheduled for delivery in April, 1981. The system will be ready for testing on October 1, 1981. Testing for performance and compliance with the ARB Rule 1135.1 will be complete in January, 1982. Further testing will continue thereafter, to evaluate catalyst life and long-term operating effects.

III. SYSTEM RETROFIT

RETROFIT REQUIREMENTS

The retrofit requirements as previously illustrated in Figures 2 and 3 for the Huntington Beach Generating Station are not at all typical for SCE generating facilities in the Los Angeles basin. Most of the larger units have horizontal shaft air preheaters which are located closely behind the boiler economizer outlet. As illustrated in Figure 5, this means that for most units all equipment and ducting downstream of the economizer outlet, including the stack, will have to be relocated to make room for the reactor and the ammonia flue gas mixer. In many cases existing structures, equipment and other site restraints interfere with the required expansion of the backend of the boiler, and consequently dictate major site rearrangements.

OPERATING AND MAINTENANCE REQUIREMENTS

Table III tabulates the O&M requirements for all Los Angeles basin units involved, including major items such as electrical power consumption, system NH_3 consumption and the yearly new catalyst and used catalyst disposal requirements. Maintenance items such as the washing and cleaning of reactor, air preheater and reactor booster fan is not known and not included at this time.

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PROBLEM AREAS

Since most Steam Generators require a major modification to the back-end of the unit in order to provide room for the installation of the reactor, retrofitting these units will require a long construction period and extended unit outages. In addition, the existing plants are already congested, which will add to this problem and create many construction restraints. Because of these problem areas and the existing unit overhaul schedule to work with, considerable risks will be encountered in meeting these schedules and supplying power to the consumers.

Some unknowns which may create further problems are catalyst availability and catalyst disposal after use. At present there are insufficient manufacturing facilities available for the new honeycomb DeNOx catalyst. The type of disposal required is not yet known nor the potential for reconditioning and reuse.

Perhaps the greatest risk to the system reliability waits at the completion of construction when startup and operation begin. Because of the booster fan required to overcome the pressure drop in the system, the larger units will be converted from forced draft units to a balanced draft operation. Controls to operate in this mode will be included and complete boiler implosion studies will be made to assess the need for boiler reinforcement. Mitigation of this risk is possible, elimination is not.

IV. COST

HUNTINGTON BEACH DEMONSTRATION UNIT

Capital Cost

Capital cost estimates have been prepared for the 107.5 MW Demonstration Project at Edison's Huntington Beach Generating Station. This estimate is provided for information, and caution should be exercised in using the figures for other applications. Since this is a demonstration project, some shortcuts have been assumed which would not be used in a commercial installation. In addition, due to the small scale of the demonstration, many of the modifications required to backfit an entire unit were not required.

The cost for this facility is estimated to be $13,500,000 including all corporate overheads and AFUDC, but excluding testing cost. This is based on a late 1981 operating date and the costs are consistent with 1981 dollars.

O&M Cost

Estimates for operating labor and material for this project were obtained from Edison's Power Supply Department. The Engineering Department provided data for ammonia, energy and catalyst consumption which became the remaining components of O&M. Catalyst replacement was calculated based on the bids received for the project. These costs are shown in Table IV.

SYSTEM RETROFIT COST

Capital

Capital cost estimates for Catalytic DeNOx were prepared in May, 1978. The basis for these estimates follows:

Conceptual engineering was performed for providing catalytic DeNOx equipment at the following units:

Units	Unit Size (Each)
Alamitos 1 & 2	175 MW
Alamitos 3 & 4	320 MW
Alamitos 5 & 6	480 MW

The engineering consisted of the development of plot plans, flow diagrams, one-line diagrams and arrangement sketches. At the completion of this effort, a list of major equipment was prepared, along with engineering take-offs of bulk material quantities such as piping, cable, structural steel, and the equipment relocations and site modifications required. Using this information, cost estimates were prepared for the above units. This information was then used to estimate the cost for all other stations using \$/kW figures from the base estimates multiplied by a site factor to account for specific conditions at each station.

Consideration was given to the following criteria in arriving at the site factor for the units listed in Table II.

Space Availability
Unit Configuration
Size of Unit

The estimates were reviewed in December, 1979, to determine if there were any inconsistencies with information collected on the catalyst and the balance of plant for the Catalytic DeNOx Demonstrations Project. It was determined that no modifications to the total costs were required. The estimates were then escalated from 1978 to 1981. These capital costs are shown in Table V.

O&M Cost

The system O&M cost was based on $/MW/year from the Catalytic DeNOx Demonstration Project modified for the unit capacity factor.

For catalyst replacement, the cost per cubic foot of catalyst of the demonstration unit was used with the assumption that the catalytic reactor volume would be proportional to the flue gas flow to be treated.

The O&M as well as catalyst replacement costs are shown in Table V.

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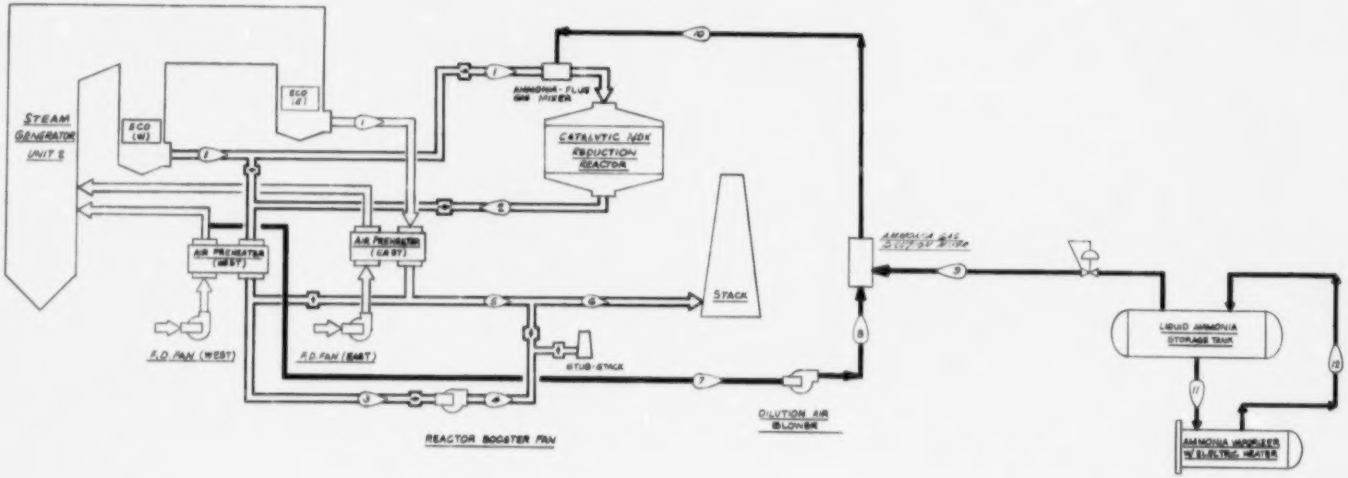

STEAM GENERATOR UNIT 2
ECO (W)
ECO (E)
AMMONIA - FLUE GAS MIXER
CATALYTIC NOX REDUCTION REACTOR
AIR PREHEATER (WEST)
AIR PREHEATER (EAST)
F.D. FAN (WEST)
F.D. FAN (EAST)
STUB-STACK
STACK
REACTOR BOOSTER FAN
DILUTION AIR BLOWER
AMMONIA GAS SHUT-OFF VALVE
LIQUID AMMONIA STORAGE TANK
AMMONIA VAPORIZER W/ ELECTRIC HEATER
1
2
3
4
5
6
7
8
9
10
11
12

	HOT FLUE GAS	TREATED HOT FLUE GAS	TREATED COLD FLUE GAS	REACTOR BOOSTER FAN DISCH.	UNTREATED COLD FLUE GAS	FLUE GAS TO STACK	DILUTION AIR SUPPLY (NOTE 3)	DILUTION AIR DISCHARGE (NOTE 3)	AMMONIA GAS (NOTE 2)	DILUTED AMMONIA GAS	LIQUID AMMONIA (NOTE 2)	AMMONIA GAS (NOTE 2)
STR NO.	1	2	3	4	5	6	7	8	9	10	11	12
Nm^3/HR.	349,150	350,840	386,610	386,610	372,810	759,420	1,610	1,610	80	1,690	—	80
SCFM	217,000	218,060	240,290	240,290	231,700	471,990	1,000	1,000	50	1,050	—	50
TEMP. °F (°C)	745 (396)	745 (396)	315 (157)	316 (157)	300 (149)	305 (149)	600 (316)	600 (316)	70 (21)	600 (316)	70 (21)	70 (21)
PRESS. "WG (PSIG) (NOTE 4)	4.5 (0.16)	-10 (-0.36)	-18 (-0.66)	2 (0.07)	2 (0.07)	1 (0.036)	13 (0.47)	40 (1.45)	(30) PSIG	40 (1.45)	(114) PSIG	(114) PSIG
ACFM	800,000	518,000	371,300	356,910	345,690	702,580	1,976	1,886	17	1,934	0.44 GPM	5.8
LB/HR.	984,000	990,714	1,092,714	1,092,714	1,053,500	2,146,214	4,585	4,585	129	4,714	129	129
NO - LB/HR (NOTE 5)	227	22.7	22.7	22.7	227	249.7	—	—	—	—	—	—
NO_x - PPM (NOTE 1)	272	27	27	27	272	150	—	—	—	—	—	—
NH_3 - LB/HR	0	5	5	5	0	5	—	—	—	—	—	—
NH_3 - PPM (NOTE 1)	0	10	10	10	0	5.8	—	—	—	—	—	—

NOTES
1 @ 3% O_2, DRY
2 NH_3 / NOX MOLE RATIO = 1
3 AIR/AMMONIA RATIO = 98 VOL %/ 2 VOL %
4. PRESSURE LEVELS ARE BASED ON:
- 22" WG F.D. FAN DISCHARGE PRESSURE
- 13.5" WG REACTOR SYSTEM AND NEW BREECHING PRESSURE DROP

5. EXPRESSED AS NO

NO.	REVISIONS
0	ISSUED FOR CONSTRUCTION
C	REVISED & ISSUED FOR DESIGN REVIEW
B	ADDED BOOSTER FAN & STUB STACK
A	PRELIMINARY ISSUE FOR REVIEW AND COMMENT

REFERENCE DRAWINGS

LOCATION HUNTINGTON BEACH STEAM STATION

CATALYTIC DENOX DEMONSTRATION
PLANT PROCESS FLOW DIAGRAM
100% LOAD—FUEL OIL

EDISON

SHEET NO. 1 of 1 SHTS.

5155701 -0

FIGURE 1

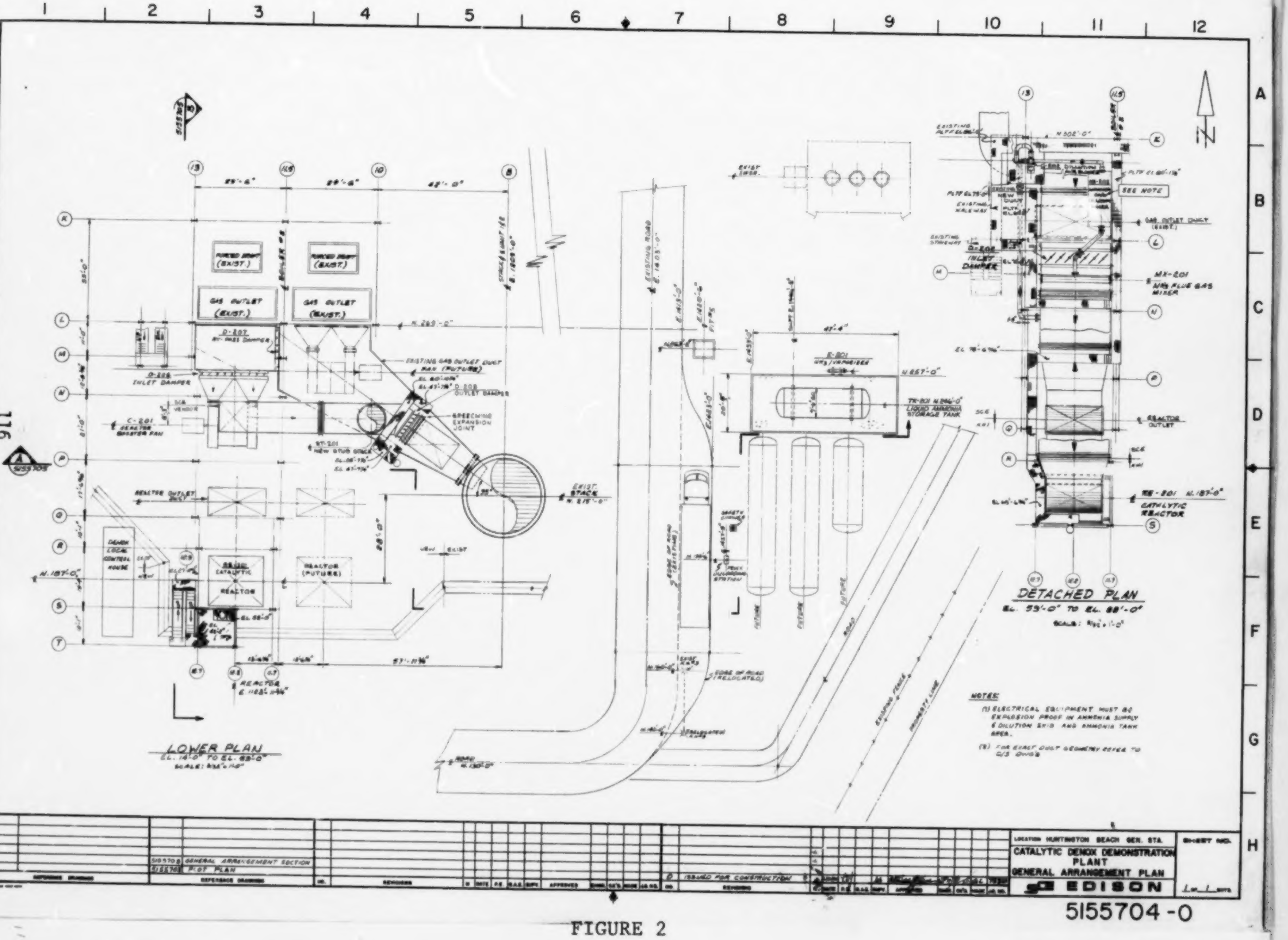
1
2
3
4
5
6
7
8
9
10
11
12
A
B
C
D
E
F
G
H
LOWER PLAN
EL. 14'-0" TO EL. 58'-0"
SCALE: 3/32" = 1'-0"
FORCED DRAFT (EXIST.)
GAS OUTLET (EXIST.)
D-207 BY-PASS DAMPER
D-206 INLET DAMPER
C-201 REACTOR BOOSTER FAN
REACTOR OUTLET DUCT
CATALYTIC REACTOR
REACTOR (FUTURE)
EXISTING GAS OUTLET DUCT FAN (FUTURE)
D-208 OUTLET DAMPER
BREECHING EXPANSION JOINT
ST-201 NEW STUB STACK
EXIST. STACK
N. 269'-0"
29'-6"
29'-6"
42'-0"
28'-0"
57'-11¾"
DENOX LOCAL CONTROL HOUSE
N. 187'-0"
REACTOR
EXISTING ROAD
EDGE OF ROAD (EXISTING)
EDGE OF ROAD (RELOCATED)
ROAD
EXISTING FENCE
PROPERTY LINE
E-201 NH₃ VAPORIZER
N. 657'-0"
42'-4"
TK-201 LIQUID AMMONIA STORAGE TANK
FUTURE
SAFETY SHOWER
AMMONIA UNLOADING STATION
EXIST. SHOP
DETACHED PLAN
EL. 59'-0" TO EL. 88'-0"
SCALE: 3/32" = 1'-0"
EXISTING PLTF ELEV.
N. 302'-0"
DILUTION AIR BLOWER
PLTF EL. 60'-1½"
SEE NOTE
GAS OUTLET DUCT (EXIST.)
EXISTING WALKWAY
EXISTING STAIRWAY
D-205 INLET DAMPER
NEW DUCT
MX-201 NH₃ FLUE GAS MIXER
EL 78'-6¾"
REACTOR OUTLET
RE-201 N. 187'-0"
CATALYTIC REACTOR
NOTES:
(1) ELECTRICAL EQUIPMENT MUST BE EXPLOSION PROOF IN AMMONIA SUPPLY & DILUTION SKID AND AMMONIA TANK AREA.
(2) FOR EXACT DUCT GEOMETRY REFER TO C/S DWG'S
5155706 GENERAL ARRANGEMENT SECTION
5155708 PLOT PLAN
REFERENCE DRAWINGS
REVISIONS
0 ISSUED FOR CONSTRUCTION
LOCATION HUNTINGTON BEACH GEN. STA.
CATALYTIC DENOX DEMONSTRATION PLANT
GENERAL ARRANGEMENT PLAN
EDISON
SHEET NO.
5155704-0

FIGURE 2

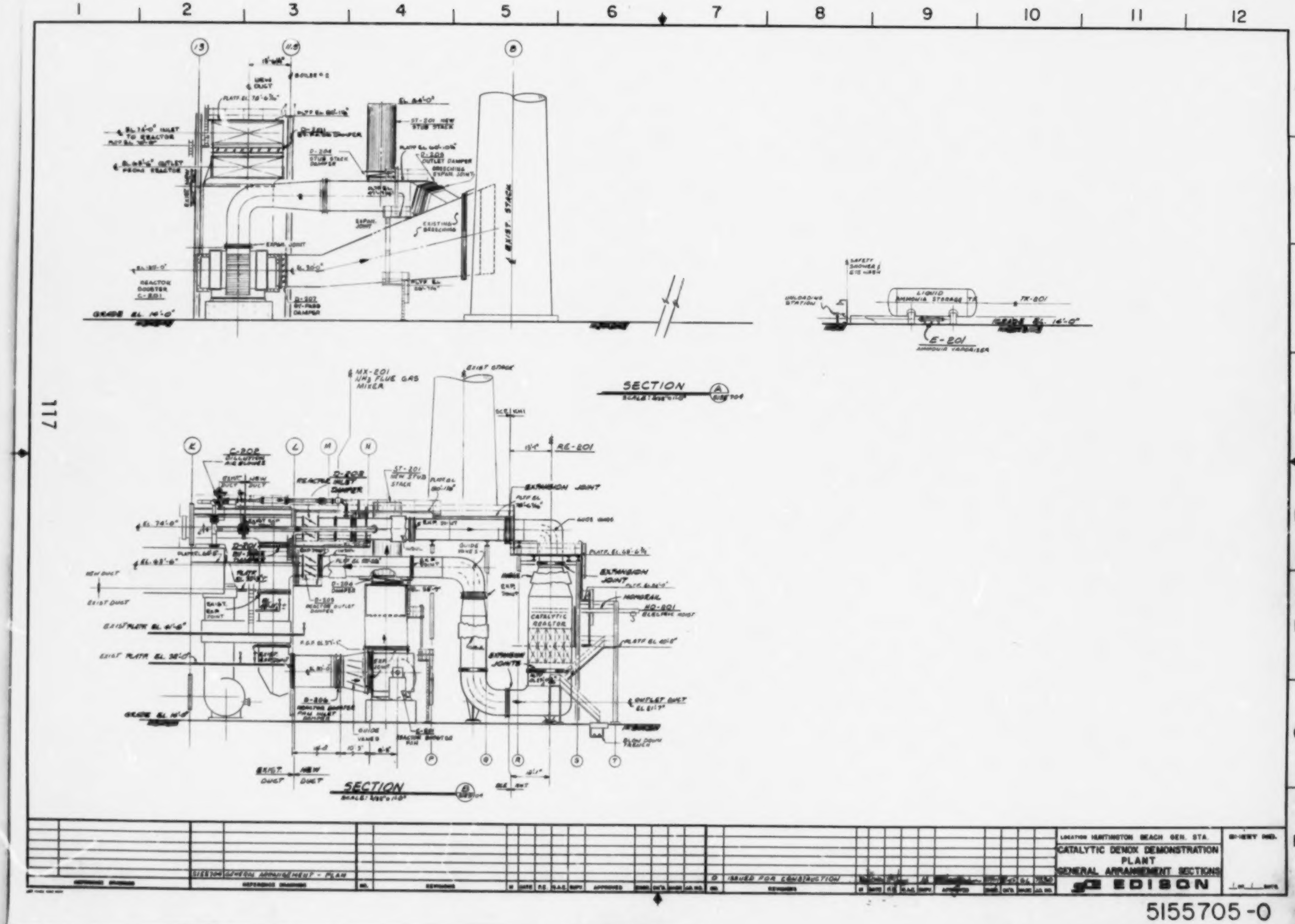

SECTION A
SECTION B
EXIST. STACK
ST-201 NEW STUB STACK
D-201 BY-PASS DAMPER
D-204 STUB STACK DAMPER
D-205 OUTLET DAMPER
BREECHING EXPAN. JOINT
EXISTING BREECHING
EXPAN. JOINT
REACTOR BOOSTER C-201
D-207 BY-PASS DAMPER
GRADE EL. 14'-0"
TO REACTOR
OUTLET FROM REACTOR
NEW DUCT
LIQUID AMMONIA STORAGE TK
TK-201
E-201 AMMONIA VAPORIZER
UNLOADING STATION
SAFETY SHOWER & EYE WASH
MX-201 NH3 FLUE GAS MIXER
EXIST STACK
C-202 DILUTION AIR BLOWER
D-202 REACTOR INLET DAMPER
RE-201
EXPANSION JOINT
EXPANSION JOINTS
CATALYTIC REACTOR
MONORAIL
HO-201 ELECTRIC HOIST
OUTLET DUCT
GUIDE VANES
D-206 REACTOR BOOSTER FAN INLET DAMPER
C-201 REACTOR BOOSTER FAN
D-203 REACTOR OUTLET DAMPER
BLOW DOWN TRENCH
EXIST. DUCT
NEW DUCT
EXIST PLATF. EL. 41'-6"
EXIST PLATF. EL. 38'-0"
EL. 74'-0"
GRADE EL. 14'-0"
ISSUED FOR CONSTRUCTION
GENERAL ARRANGEMENT - PLAN
LOCATION HUNTINGTON BEACH GEN. STA.
CATALYTIC DENOX DEMONSTRATION PLANT
GENERAL ARRANGEMENT SECTIONS
EDISON
SHEET NO.
5155705-0

FIGURE 3

O 129A NEW 8 70
S.C.E. CO.

PLANNING SCHEDULE

GENERATING STATION: Huntington Beach
UNIT NO: 2
DATE: 7-29-80

MAJOR ACTIVITIES FOR CATALYTIC DeNOx PROJECT

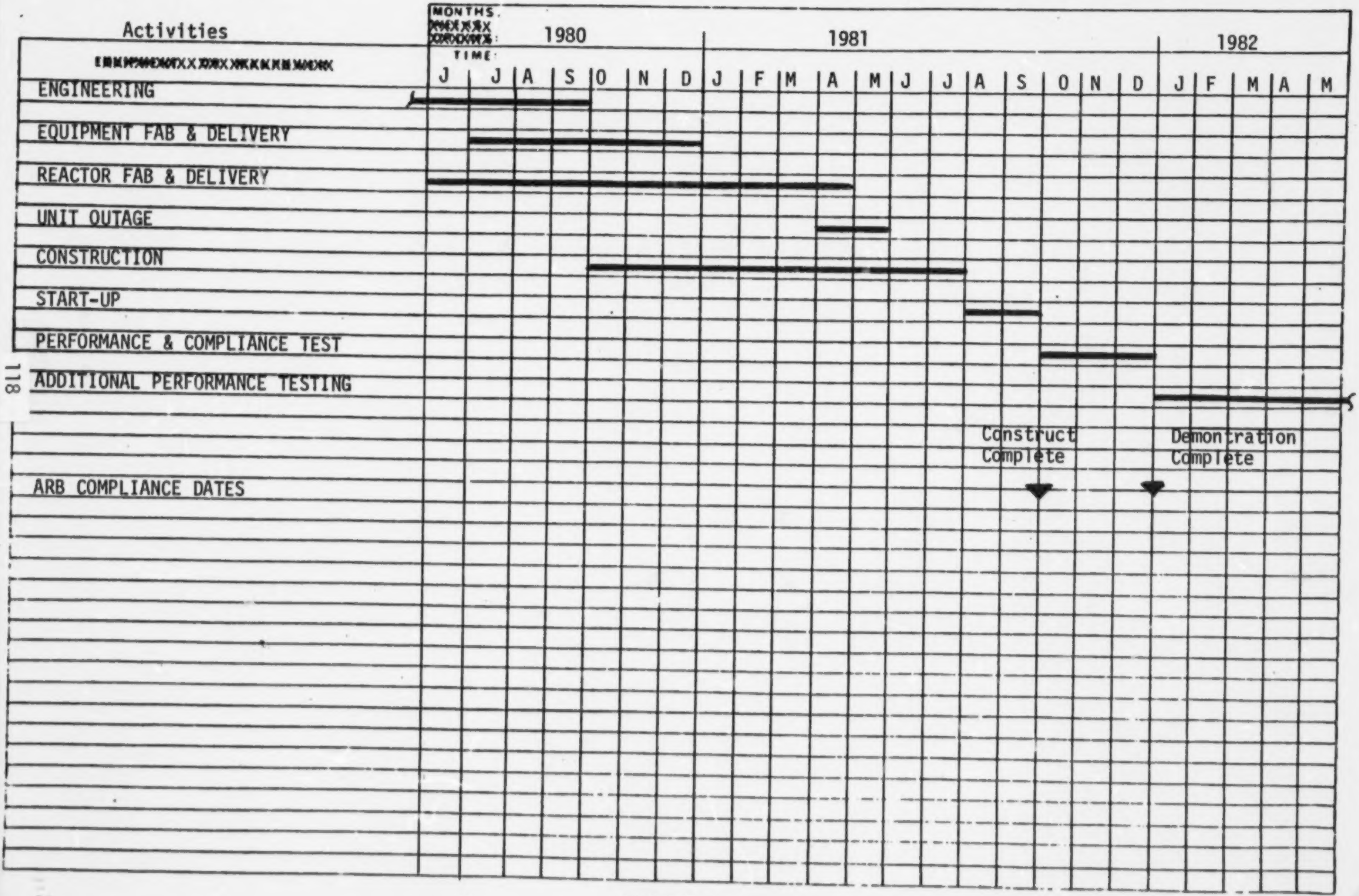
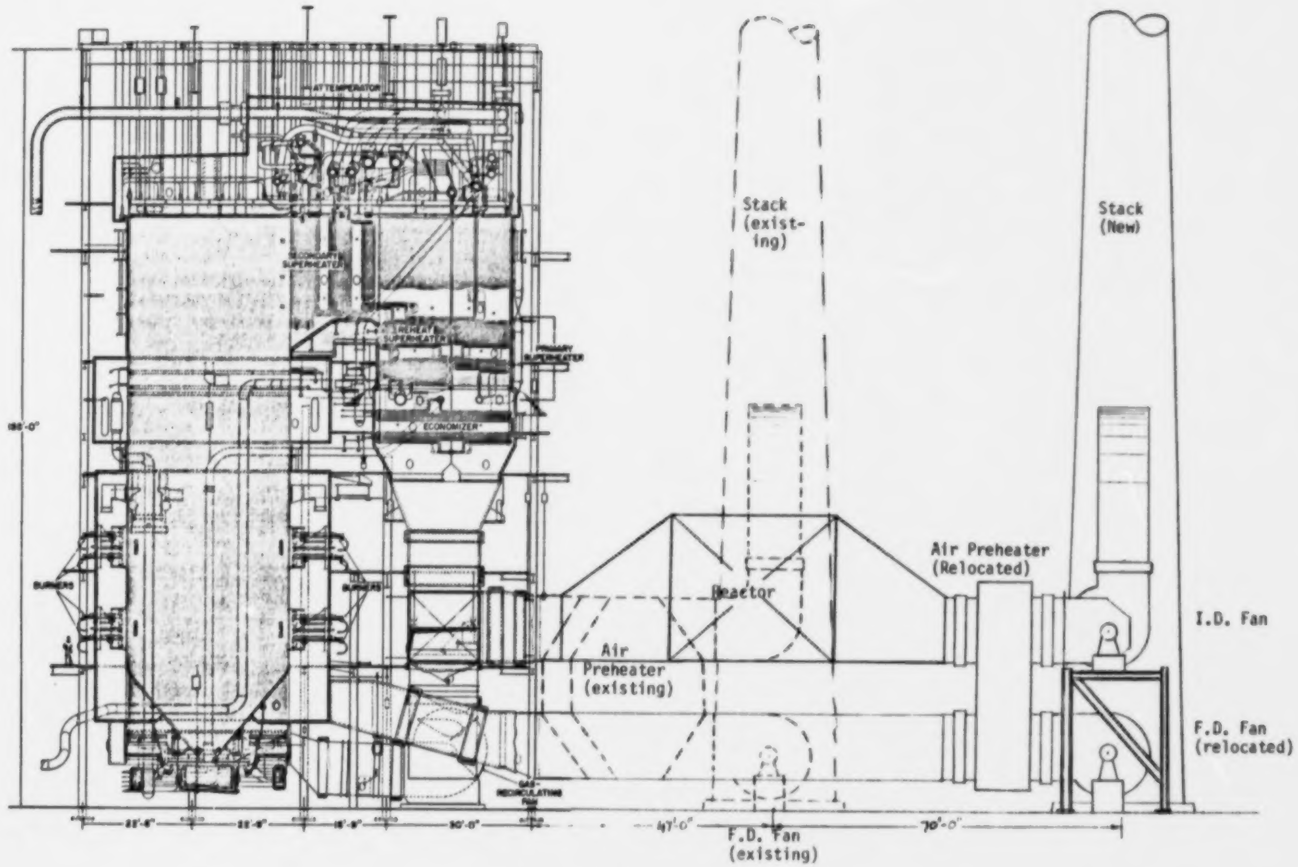


FIGURE 4


ATTEMPERATOR
SECONDARY SUPERHEATER
REHEAT SUPERHEATER
PRIMARY SUPERHEATER
ECONOMIZER
BURNERS
BURNERS
GAS-RECIRCULATING FAN
Stack (exist-ing)
Stack (New)
Reactor
Air Preheater (existing)
Air Preheater (Relocated)
I.D. Fan
F.D. Fan (relocated)
F.D. Fan (existing)
47'-0"
70'-0"

SOUTHERN CALIFORNIA EDISON COMPANY
ALAMITOS STEAM STATION - UNITS NO. 5 & 6
LOS ALAMITOS, CALIFORNIA

SHOWN RETROFITTED WITH SCR NOx REMOVAL REACTOR

FIGURE 5

TABLE I

HUNTINGTON BEACH UNIT 2 DATA

- Electrical Output: 215 MW (107.5 MW for West half of unit)
- Flue Gas Flow Rate: 100,000 to 500,000 ACFM at 750°F (half of total unit flow)
- Flue Gas Temperature: 550° to 800°F (economizer outlet)
- NOx Concentration: 180-280 vppm at 3% O_2 dry
- SO_2 Concentration: 70-140 vppm at 11% and 4% O_2
- Particulate Concentration: .005 - .02 gr/SCF
- SO_3 Concentration: 1 - 5 vppm

CO:df

TABLE II

MAJOR EQUIPMENT DATA

Catalytic Reactor:

- Size 16′ (W) x 18′ (L) x 23′ (H)
- Space Velocity: 6655 hr^{-1}
- Design Pressure: -17 to +15" WG
- Assembled Weight: 184,300 lbs
- Catalyst Shape: Square Honeycomb

Booster Fan:

- Type: centrifugal, air foil blading
- Operating characteristics: 371,800 CFM @ 17" WG SP.
- Flow control: inlet vanes
- Drive: 2000 HP Electric Motor

Ammonia Tank:

- Nominal Capacity: 12,000 gallon

Electrical Equipment:

- 4160 V Switchgear
- 480 V Circuit Breaker
- 480 V Motor Control Center

CO:df

TABLE III

OPERATING AND MAINTENANCE DATA

Huntington Beach SCR Demonstration Unit (107.5 MW)

o Power Requirement	1500 kW
o Power Consumption	7.5×10^6 kWhr/yr
o NH_3 Consumption	320 tons/yr
o Catalyst Consumption* and Disposal	20 tons/yr

SCR NOx Removal Systems on all 175 MW and larger L.A. Basin Units (7720 MW)

o Power Requirements	83 MW
o Power Consumption	386×10^6 kWhr/yr
o NH_3 Consumption	21,500 tons/yr
o Catalyst Consumption* and Disposal	1,860 tons/yr

*Based on two-year catalyst operating life

CO:df

TABLE IV

107.5 MW

CATALYTIC DeNOx - DEMONSTRATION UNIT

O&M COST (1981 $)

Derate	$ 59,880
Energy	207,600
Chemicals	62,570
Labor and Supplies	134,250
Catalyst Replacement	986,740
Total O&M	$1,362,860
SAY	$1,363,000

CO:df

TABLE V

COST SUMMARY FOR CATALYTIC DeNOx REDUCTION TECHNIQUES

($ X 10^6 all in 1981)

	Total Capacity (MW)	Average(1) Capacity Factor (%)	Capital ($)	Catalytic DeNOx(2)		
				O & M (1-Year) ($)	Catalyst (1-Year Life) ($)	Replacement(3) (2-Year Life) ($)
Ormond 1 & 2	1500	55	220.0	7.2	10.1	5.1
Redondo 7 & 8	960	59	190.0	4.9	6.4	3.3
Alamitos 5 & 6	960	59	130.0	4.9	6.4	3.3
Alamitos 3 & 4	640	53	80.0	3.0	4.1	2.1
El Segundo 3 & 4	670	53	120.0	3.2	4.5	2.3
Etiwanda 3 & 4	640	53	100.0	3.0	4.1	2.1
Huntington 3 & 4	440	47	60.0	2.0	2.8	1.4
Huntington 1 & 2	430	47	60.0	1.8	2.7	1.4
Mandalay 1 & 2	430	47	60.0	1.8	2.7	1.4
Alamitos 1 & 2	350	40	50.0	1.4	2.4	1.2
El Segundo 1 & 2	350	40	50.0	1.4	2.4	1.2
Redondo 5 & 6	350	40	60.0	1.4	2.4	1.2
Total	7720		1180.0	36.0	51.0	26.0

NOTE: (1) Average capacity factor - Based on the latest information from System Planning - November 1979. These factors were used in the calculation of O&M.

(2) Catalytic DeNOx - The capital cost was based on the original estimate of May 1978. The O&M cost was based on the latest information available from the demonstration unit in Huntington Beach Station and also using the latest capacity factors available.

(3) Catalyst Replacement - Cost shown is an average cost for a 2-year life catalyst.

CO:df

COUNTERMEASURES FOR PROBLEMS IN NO_x REMOVAL PROCESS FOR COAL-FIRED BOILERS

By:

H. Itoh and Y. Kajibata
Kawasaki Heavy Industries, Ltd.
Japan

COUNTERMEASURES FOR PROBLEMS IN NOx REMOVAL PROCESS FOR COAL-FIRED BOILER

Hayami Itoh
Kawasaki Heavy Industries, Ltd.
Japan

Takahiro Kajibata
Kawasaki Heavy Industries, Ltd.
Japan

ABSTRACT

The construction of many coal-fired power plants is being planned as a result of the recent petroleum shortage , and so the need for the De-NOx process for coal-fired boilers is increasing. However, there are many problems to be solved in the practical application of the De-NOx process for coal-fired boilers, because high concentrations of dust particles and SOx are contained in the flue gas. The major problems are : (1) catalyst bed pluggage and catalyst erosion by dust particles (2) the influence of unreacted NH_3 and SO_3 from the De-NOx reactor to the downstream equipment (3) deactivation of the catalyst by dust particles and SOx.

KHI has been working to solve these problems and to put the De-NOx process into practical use for coal-fired boilers with the co-operation of EPDC for many years. We have developed superior catalysts having several characteristics, which include long life, SOx resisting properties, dust resisting properties and low conversion of SO_2 to SO_3. Further, we have solved the above problems and have developed the most economical and stable De-NOx process.

This paper describes the problems involved in the practical application of the De-NOx process for coal-fired boilers and the countermeasures undertaken in KHI's De-NOx process.

INTRODUCTION

It was since 1973 that the regulation on NOx emission from fixed source was enforced in Japan. Many enterprises have been involeved in research and development of NOx removal technology. Kawasaki Heavy Industries Co., Ltd. (hereinafter referred to as " KHI ") has been endeavoring to the NOx removal technology for many years, and has in particular engaged in the research and development with respect to the Selective Catalytic Reduction of NOx with NH_3 since 1970. Finally, we have succeeded in the practical application of the DeNOx process for LNG-fired and heavy oil-fired boilers.

On the other hand, the practical application of the DeNOx process for coal-fired boilers is more difficult than for oil-fired boilers, because the flue gas of coal-fired boilers contains more dust particles and SOx as compared to that of oil-fired boilers. Since 1975, KHI has been cooperating with Electric Power Development Co., Ltd. (hereinafter referred to as EPDC) in the research of DeNOx process for coal-fired boilers. We have developed superior catalysts having several characteristics, which feature long life, SOx resisting properties, dust resisting properties and low conversion of SO_2 to SO_3. Furthermore, we have obtained the economical and stable DeNOx process. Consequently,KHI received an order for demonstration DeNOx plant for Takehara No.1 boiler of EPDC and are now doing our best to manufacture it. Furthermore, KHI received an order for the DeNOx pilot plant from Electric Power Research Institute in the U.S.A. and are now engaged in its operation.

In this paper , we describe the various problems involved in practical application of the DeNOx process for coal-fired boilers and countermeasures undertaken in KHI's DeNOx process.

VARIOUS PROBLEMS IN DeNOx PROCESS FOR COAL-FIRED BOILERS

Because of high concentration of dust, SOx and NOx in the flue gas out of the coal-fired boiler, the total system is required for flue gas treatment in order to resolve the environmental problems. The total flue gas treatment system is composed of dust collection, DeNOx and DeSOx. In order to improve the performance, reliability and economics of the total system, we must consider not only the performance and reliability of each component technic but also the most suitable combination of these components. Accordingly, the performance and reliability of the DeNOx process have to be pursued considering its influence on the other process.

There are two kinds of DeNOx processes for coal-fired boilers, viz. low dust DeNOx process and high dust DeNOx process (cf. Figure 1). We have solved two problems in developing these DeNOx processes. The first problem is the influence of the unreacted NH_3 and the generated SO_3 from the DeNOx reactor on the downstream equipment, and the second one is the influence of the high concentration of dust and SOx in the flue gas on the performance and stability of the DeNOx plant.

Influence of the unreacted NH_3 and the generated SO_3 from the DeNOx reactor on downstream equipment

Influence of the unreacted NH_3. In the low dust DeNOx process, the unreacted NH_3 reacts with SO_3 and forms ammonium bisulfate with the fall of the flue gas temperature at the air preheater. As the ammonium bisulfate is strongly corrosive and adhesive , it causes the corrosion or plugging of the air preheater. And as the unreacted NH_3 is collected in the wet DeSOx process and accumulated in the absorbing liquid, there arises a need for removing NH_3 in the waste water of the wet DeSOx process.

In the high dust DeSOx process, the unreacted NH_3 is fixed to the fly ash which is collected by the electrostatic precipitator. Figure 2 shows the relationship between the concentration of the unreacted NH_3 and the adsorbed ammonia on fly ash. The dotted line in the figure shows the calculated value assuming that all the unreacted NH_3 is adsorbed on the fly ash. According to the test results, most of the unreacted NH_3 less than 20ppm is proved to be adsorbed on the fly ash. Accordingly, the influence of the unreacted NH_3 on the air preheater and the wet DeSOx process in the high dust DeNOx process is less than in the low dust DeNOx process. But there are several problems such as leaching of NH_3 and offensive odor in utilizing the fly ash for cement admixing material or using it for reclamation.

As the unreacted NH_3 will cause various sorts of troubles,it is necessary to decrease to the unreacted NH_3 at least as low as 10ppm and preferrably below 5ppm (1).

Influence of SO_3. The DeNOx catalyst generally works as shown in the formulas

(1) and (2). In addition to these NOx removal reactions, it works as SO_2 oxidation catalyst as in the formula (3).

$$
\begin{array}{llr}
NO + NH_3 + 1/4\ O_2 & \longrightarrow N_2 + 3/2\ H_2O & (1) \\
NO_2 + 4/3\ NH_3 & \longrightarrow 7/6\ N_2 + 2\ H_2O & (2) \\
SO_2 + 1/2\ O_2 & \longrightarrow SO_3 & (3)
\end{array}
$$

In the low dust DeNOx process, SO_3 forms ammonium bisulfate through the reaction with the unreacted NH_3 and forms the sulfulic acid at low temperatures.

In the high dust DeNOx process, there is no influence of SO_3 to the air preheater, because most of SO_3 is adsorbed on the fly ash. But SO_3 adsorbed on the fly ash accelerates the adsorption of NH_3.

As SO_3 from the DeNOx reactor will cause the above mentioned troubles, it is desirable to keep the oxidation of SO_2 as low as possible. It is necessary to decrease SO_3 below 10ppm in order to control influence of SO_3 to the minimum.

Influences on the performance and stability of DeNOx process

Influence of dust. Problems of the plugging of the catalyst bed and the erosion of the catalyst are perceived as the influence of the dust. The plugging of the catalyst bed will cause the increase of the pressure drop of the catalyst bed and the decrease of the NOx removal efficiency, whereas the erosion of the catalyst will cause the decrease of the NOx removal efficiency and the secondary environmental pollution by scattering the catalyst components. Accordingly, sufficient countermeasures are required against the plugging and the erosion caused by dust, especially in the high dust DeNOx process.

Influence caused by the particular properties of coal-fired flue gas.
Influences of SOx, halogen (HCl,HF), and alkali metals in dust, especially pottassium, on the catalytic performances may be anticipated. The decrease of the catalytic performance must be especially avoid, because it causes not only the lack of stability of the system but also the increase of running cost. Accordingly, it is necessary to develop catalysts which can resist against the particular properties of coal-fired flue gas.

Influence of the long-term low load operation and the repeat of star-up/shut-down. The long-term low load operation of boilers may result in the deactivation of the catalyst and the plugging of the catalyst bed due to the deposit of NH_3 , SOx and dust in accordance with the decrease of the gas temperature and the gas flow rate.

Repeat of start-up/shut-down of boilers may result in the catalytic deactivation due to the condensation of SOx and H_2O on the catalyst surface and due to masking of the catalyst surface with dust.

It is necessary to perform the simulation tests of the long-term low load operation and the repeat of start-up/shut-down.

COUNTERMEASURES ADOPTED IN KHI's DeNOx PROCESS

Control of the unreacted NH_3 and the oxidation of SO_2

Control of the unreacted NH_3. The NH_3/NOx mole ratio should be controlled less than 1 in order to control the unreacted NH_3 emission. For example, it is necessary to control the NH_3 injection whithin the region of NH_3/NOx mole ratio between 0.83 and 0.93 in order to obtain the NOx removal efficiency more than 80% and to decrease unreacted NH_3 less than 5ppm (cf. Figure 3.).

For this purpose, it is required to control the NH_3 injection in accordance with the change of the volume of the treatment gas and that of the inlet NOx concentration. KHI developed an unique control system by combining the following two methods. One is a feed-forward control of the NH_3 injection, of which the amount is calculated by the boiler load and the inlet NOx concentration. The other is a feed-back control, in which the amount of NH_3 injection is corrected by the outlet NOx concentration or the NOx removal efficiency. The follow-up test results of our control system for boiler load variation was satisfactory. As shown in Figure 4, the unreacted NH_3 is controlled within 5ppm in spite of the change of the gas volume, the gas temperature as well as the inlet NOx concentration.

In large DeNOx plants, the mixing and dispersing of injected NH_3 are very important in order to decrease the unreacted NH_3. We carried out cold-model tests as to the mixing and dispersing, and have found the most efficient

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configulation. The length of the duct from the nozzle to the reactor is short and the pressure drop of the duct is quite small in this configulation.

Control of SO_2 oxidation. Characteristic of a conventional catalyst are shown in Figure 5. The Oxidation rate of SO_2 will increase as the NH_3/NOx mole ratio and the space velocity decrease. Therefore, the development of catalysts with low oxidation rate of SO_2 is indispensable for the simultaneous control of the unreacted NH_3 and the generated SO_3 emission. KHI has endeavored to improve the catalysts in cooperation with the catalyst manufacturer.

We started with the V_2O_5-TiO_2 basis catalyst, which has various features such as high NOx reduction activity and long life, etc..
Our points of the improvement are as follows :

1. Decrease of the amount of the active element (V_2O_5)
2. Addition of an active element as promoter
3. Improvement of the dispersion of the active elements

We succeeded in developing the catalysts with low oxidation rate of SO_2 and high NOx reduction activity. Figure 6 shows the performance of the two improved catalysts. KHI has several catalyst with low oxidation rate of SO_2 avilable , and select the most suitable catalyst for practical use in accordance with gas temperature and SOx concentration. Figure 7 shows the performance of the improved catalyst (K-110) for coal-fired boiler load conditions. Thus it is possible to control SO_3 emission from the DeNOx reactor below 10ppm by employing the improved catalysts with low oxidation rate of SO_2.

Treatment of the ash containing NH_3. As already mentioned, NH_3 is contained in the fly ash collected by the electrostatic precipitator in the high dust DeNOx process. The ash containing NH_3 being dumped for reclamation,there might arise problems of NH_3 odor and leaching of NH_3. We carried out odor tests and leaching tests, and confirmed that it is necessary to make the amount of NH_3 on the fly ash below 20ppm (mg/kg) for reclamation. We have been developing the NH_3 removal technology out of the fly ash in cooperation with EPDC. Our NH_3 removal process is a pyrolysis process by utilizing a part of flue gas of the boiler. We have carried out the bench scale tests and have confidence in the practical application of this process.

Means of stabilizing the catalystic performance

Countermeasures against dust. As already mentioned, there are two problems due to the dust, viz. the plugging of the catalyst bed and the erosion of the catalyst. Against the plugging of the catalyst bed, we have been employing fixed bed reactor with dust free type catalysts for many years. Through the pilot tests for the dust free type catalysts in high dust and low dust flue gases, we have found the most suitable catalyst shape, catalyst dimensions and gas flow rate for respective flue gas. As for the shape of the catalyst, pipe or honeycomb shape is most preferable, but for the high dust gas wider opening is required than for the low dust gas.

The erosion of the catalyst usually starts at the inlet part of the catalyst and gradually progresses (2) . The countermeasure we adopted is hardening the inlet part of the catalyst as long as 1/100 to 1/50 of its length by the impregnant, of which effectiveness was confirmed by endurance tests. Our catalysts are made so as to contain in the active elements evenly into the inside, and so the performance of the catalysts will be maintained in spite of the progress of the erosion. Figure 8 shows the change of the catalyst erosion rate with time for a conventional catalyst and our improved catalyst. The inlet hardening could decrease the amount of erosion of about 6% per year to less than 1%.

Long-term stability of the catalytic performence. KHI's catalysts have various features such as high activity and long life. In our previous report, we had confirmed that the lifetime of the catalyst for the flue gas of the heavy oil-fired boilers was more than two years (2) . For coal-fired boilers, we carried out the durability test at the pilot plants adopted to Takehara No.1 boiler of EPDC and could confirm that the lifetime of the catalyst was more than 8,000 hours. As shown in Figure 9, no change was noticed of NOx removal efficiency as well as the pressure drop due to the catalyst bed. For the improved catalyst with low oxidation rate of SO_2, the durability test has been continued for more than 3,000 hours. According to Figure 10, no change is noticed of the NOx removal efficiency and of the oxidation rate of SO_2. As a result, it was confirmed that our catalyst has a stable performance against SOx, halogen (HCl, HF) in the flue gas and potassium in the dust.

In order to investigate the influence of the long-term low load operation and the repeat of start-up/shut-down of the boiler, we conducted simulation tests. As for the long-term low load operation, we conducted cyclic operations between a quarter and a half of the full load for about one month. It was found that the catalytic activity could be recovered by the full load operation at 350°C, although it was temporarily lowered by the low load operation at 270°C (cf. Figure 11). This temporary deactivation phenomenon is consider to be caused by the increase of SO_4^{2-} and NH_4^+ in the catalyst. But the catalyst will not reach the permanent poisoning by such a degree of increase of SO_4^{2-} and NH_4^+ . NH_4^+ is removed from the catalyst with the recovery of reaction temperature, and the catalytic activity is recovered. On the other hand, we repeated emergency stop and start-up five times. No deactivation of the catalyst was caused by such operations like as emergency stop and start-up (cf. Figure 11) .

KHI's DeNOx PROCESS FOR COAL-FIRED BOILERS

The KHI's newly developed DeNOx process for coal-fired boilers has the following features :

1. It is possible to suppress the unreacted NH_3 below 5ppm by means of our highly active catalysts, pertinent control of NH_3 injection and uniform dispersion of NH_3 .
2. It is possible to control the SO_3 from the DeNOx reactor below 10ppm by employing the catalysts of low oxidation rate of SO_2 .
3. No plugging of the catalyst bed occurs by employing such dust free type catalyst as pipe-shaped or honeycomb-shaped.
4. The erosion of the catalyst will hardly occur even in the high dust DeNOx process by employing the hardening treatment of the inlet part.
5. Our catalysts are hardly subject to the influence of SOx, halogen in the flue gas and potassium in the dust .

In addition to this DeNOx process, KHI has the established technology of dust collection and wet DeSOx , and has been proceeding with the development of the comprehensive flue gas treatment system for coal-fired boilers. The flow diagram of the toatl flue gas treatment systems and an example of material balance for a 500 MW coal-fired boiler are shown in Figures 12 and 13. As shown in these figures, it can be seen that the unreacted NH_3 is suppressed to 5ppm and the generated SO_3 is to 8ppm with the NOx removal efficiency as high as 90% . And it can be seen that unreacted NH_3 and generated SO_3 are all collected by the electrostatic precipitator in the high dust DeNOx process, and are partially collected by the wet DeSOx process in the low dust DeNOx process.

SUMMARY

Hereto described is the progress to date of the KHI's DeNOx process with many outstanding features for coal-fired boilers. KHI has almost accomplished the total system for flue gas treatment utiliziling this process. However, further study tasks are continuing for improvement of reliability and economics of the total systems.

Finally, we wish to express our sincere appreciation to Mr. Nakabayashi, Mr. Shimizu and Mr. Mohri of Electric Power Development Co., Ltd..

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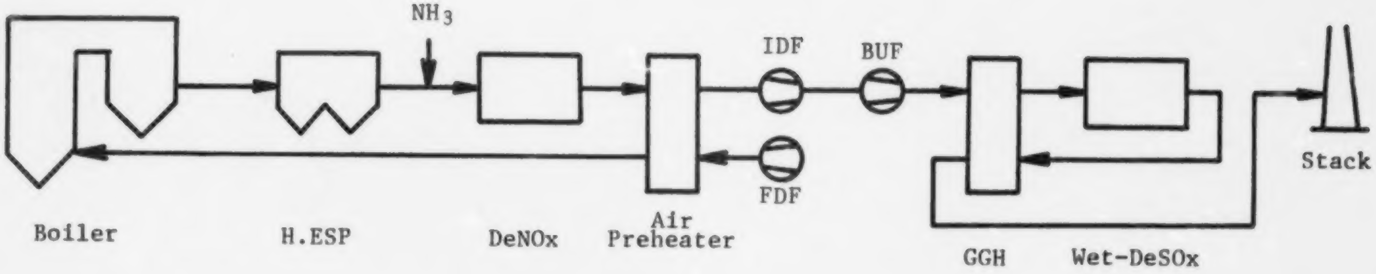
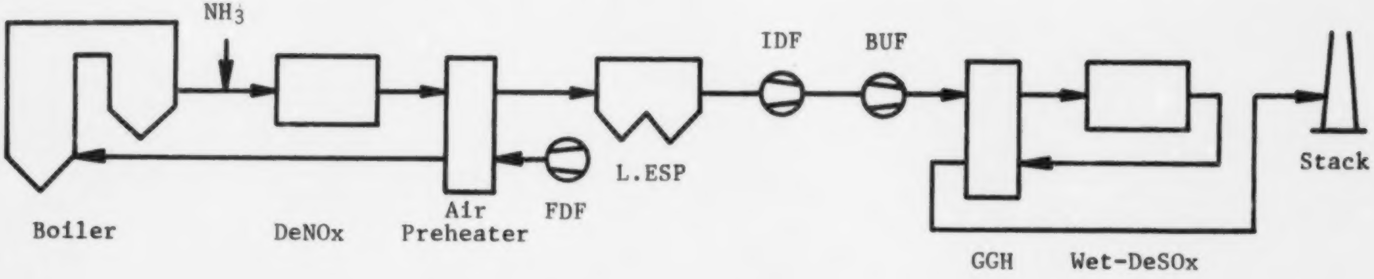
2. Y. Nakabayashi and S. Niwa, " Characteristics of Cylindrical De-NOx Catalysts for a Coal-fired Boiler ", EPRI FP-1109-SR Seminar Proceedings Special Report, pp21-1 – 21-13, July 1979


NH3
IDF
BUF
FDF
Stack
Boiler
H.ESP
DeNOx
Air Preheater
GGH
Wet-DeSOx

Low Dust DeNOx Process


NH3
IDF
BUF
Stack
L.ESP
Boiler
DeNOx
Air Preheater
FDF
GGH
Wet-DeSOx

High Dust DeNOx Process

Figure 1. Total Flue Gas Treatment Process for Coal-Fired Boilers

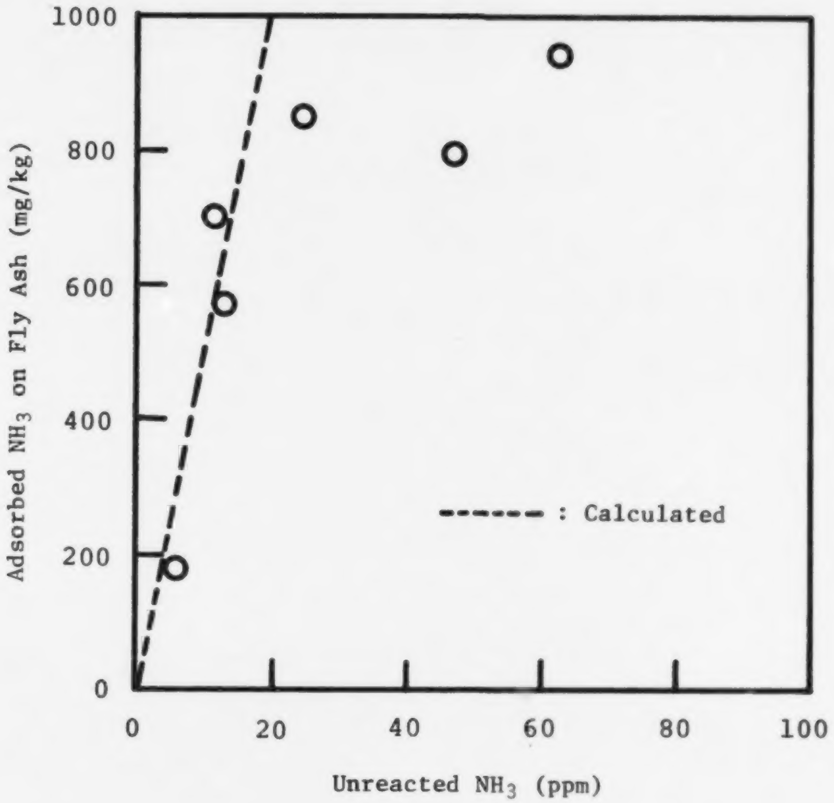


Figure 2. Adsorption of Unreacted NH_3 on Fly Ash

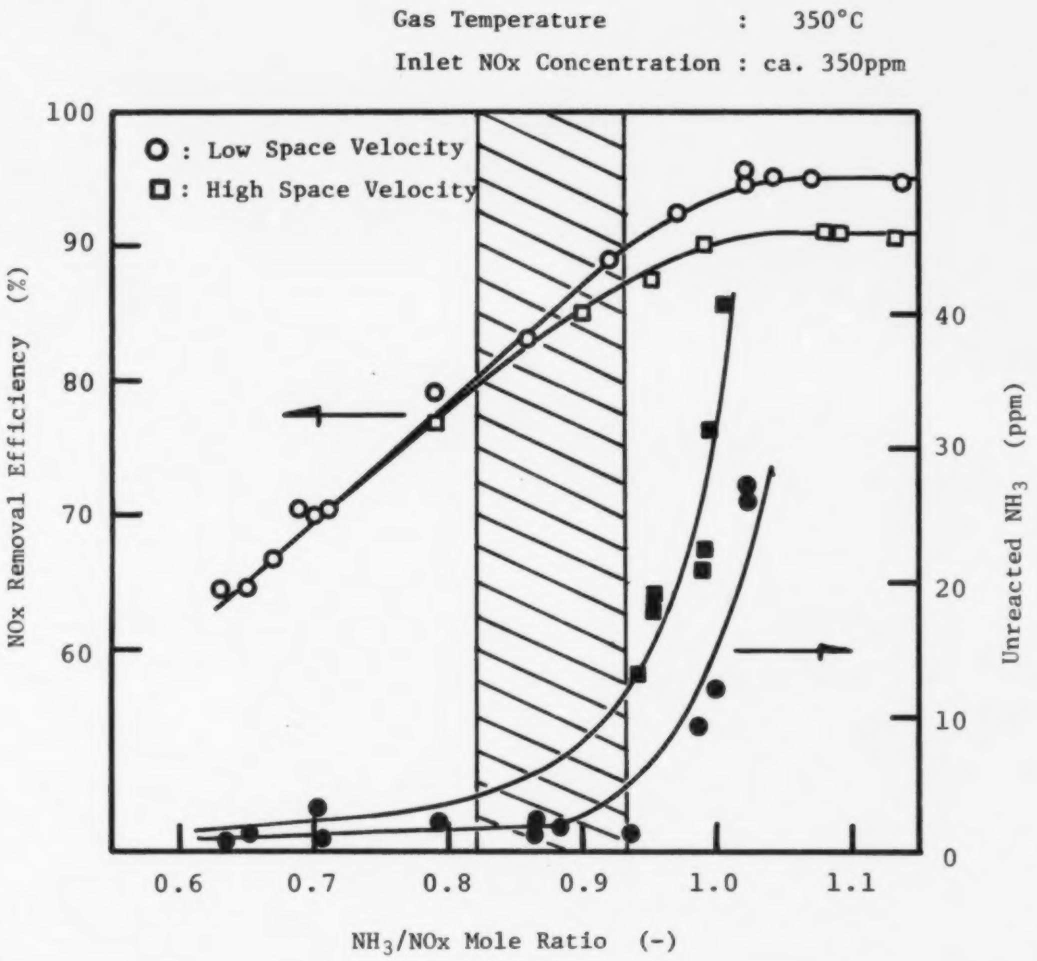


Figure 3. Control Region of NH_3/NOx Mole Ratio

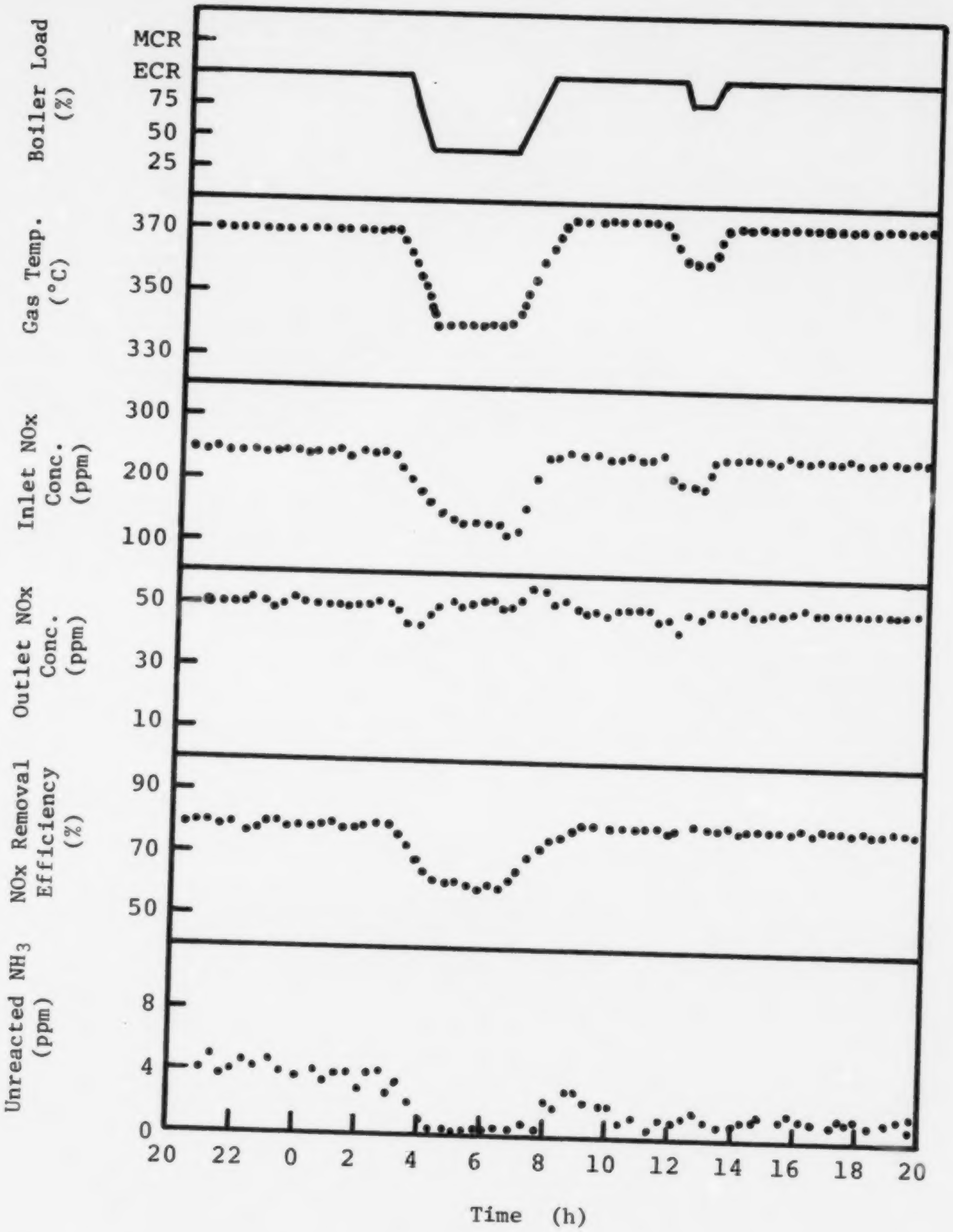


Figure 4. Follow-up Test for Boiler-Load Variation

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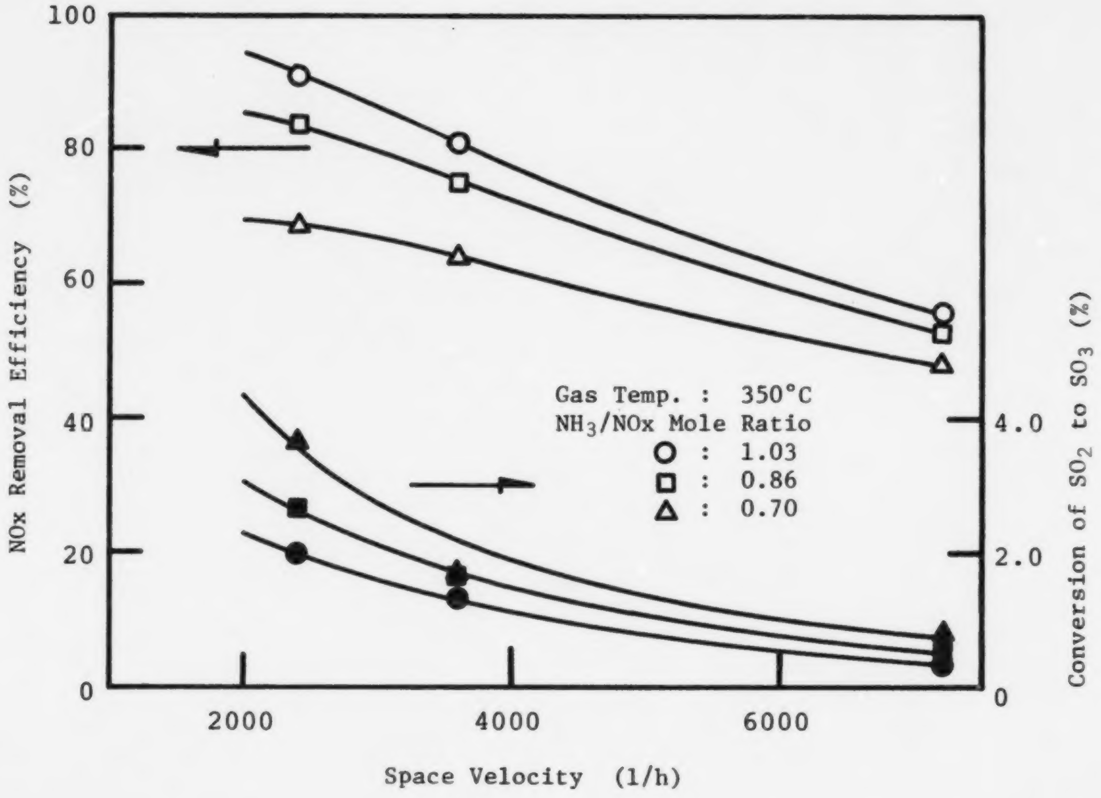


Figure 5. Characteristic of a Conventional Catalyst

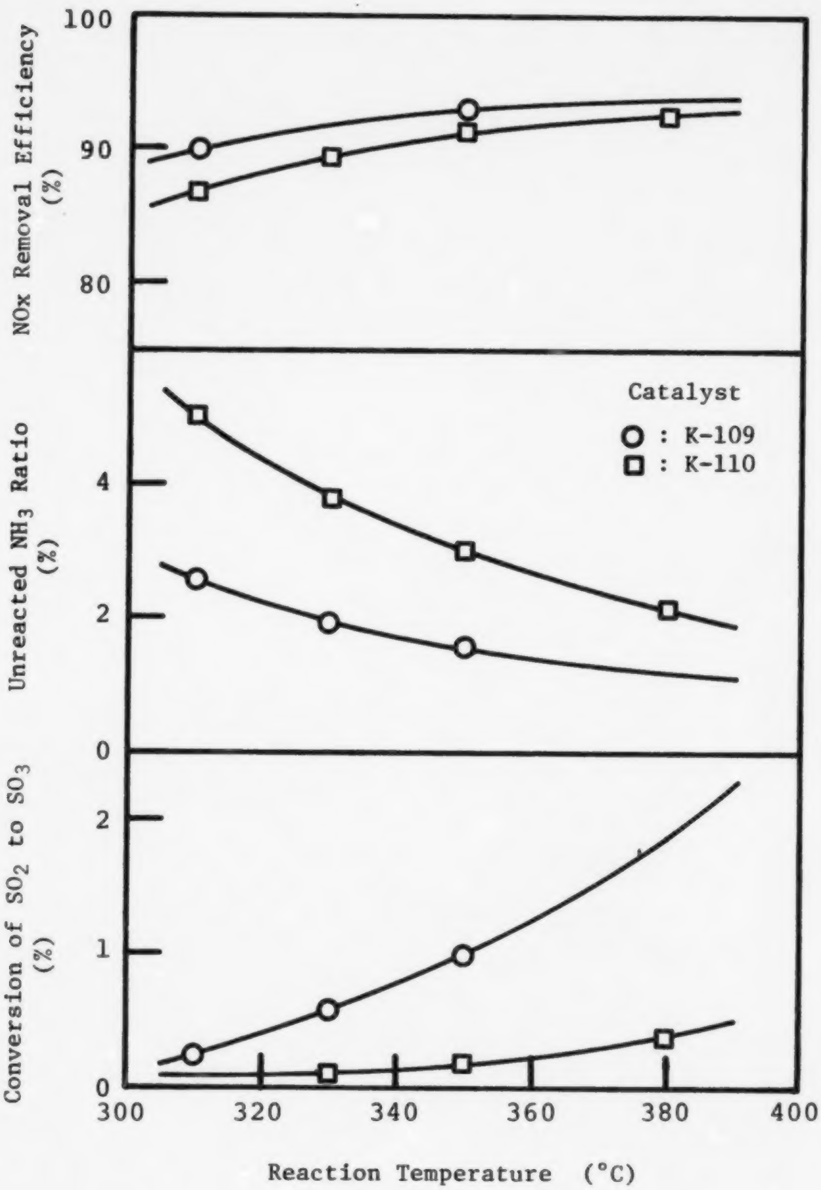


Figure 6. Performance of Improved Catalysts

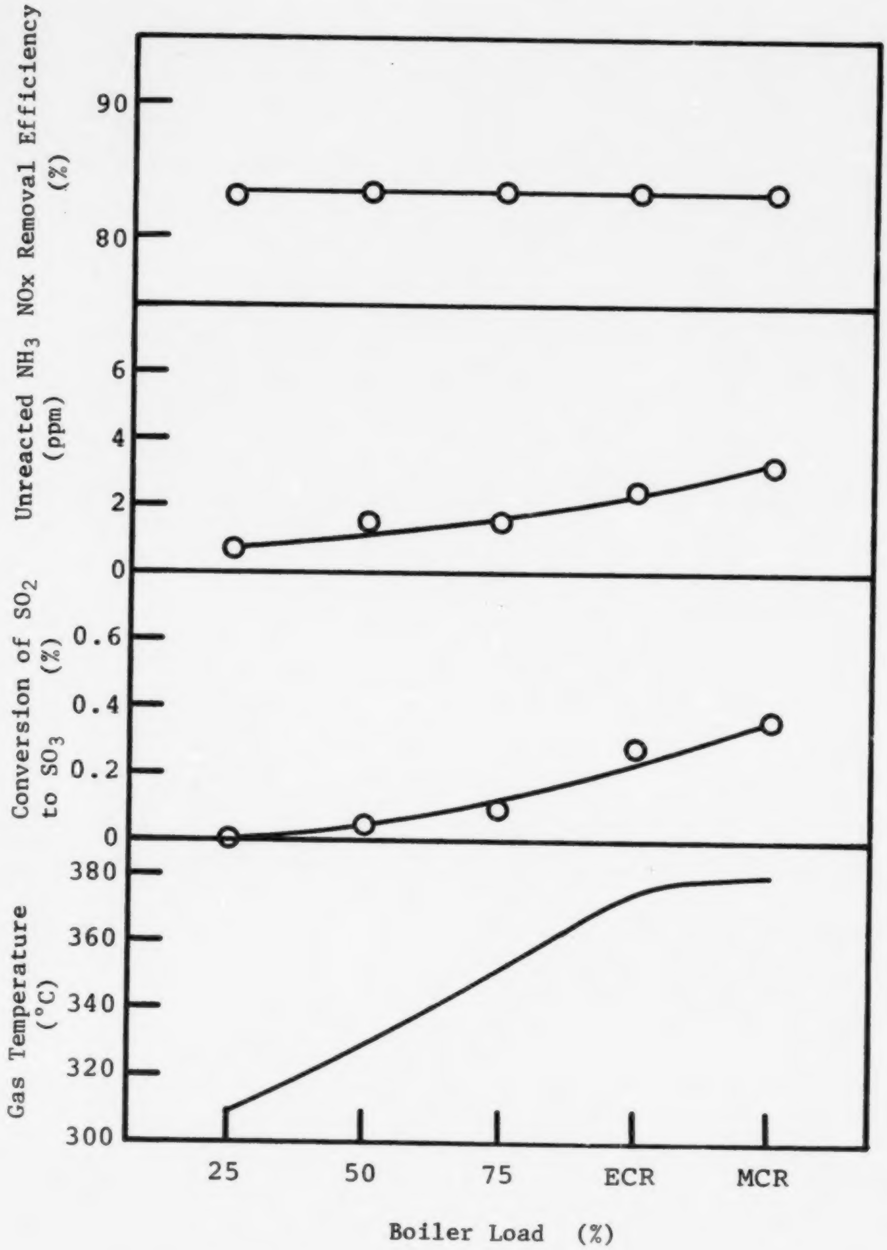


Figure 7. Performance of Improved Catalyst (K-110) for Boiler Load Conditions

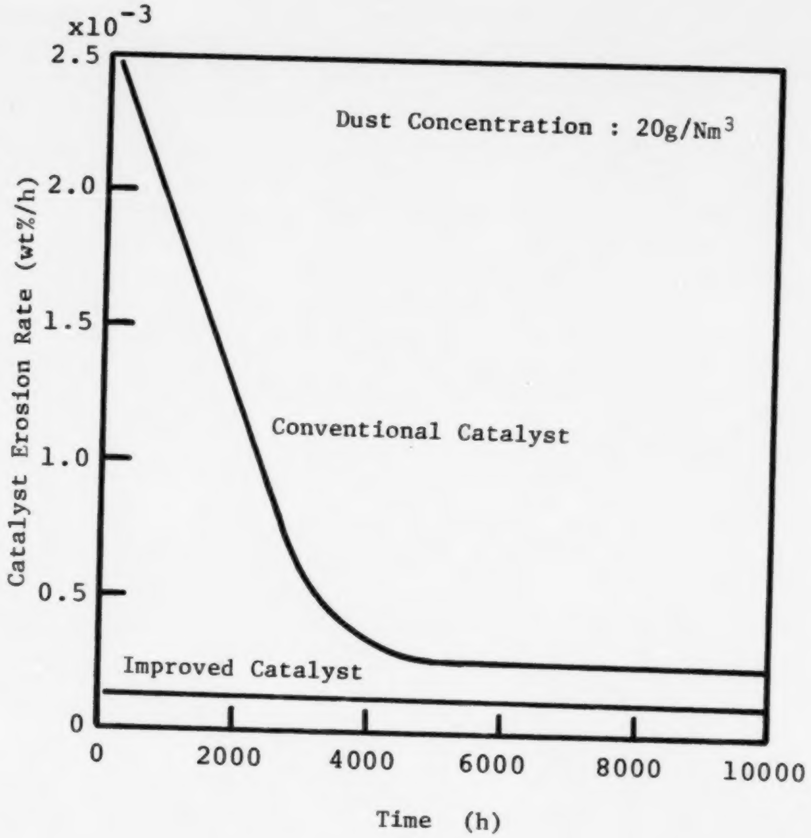


Figure 8. Change of Catalyst Erosion Rate with Time

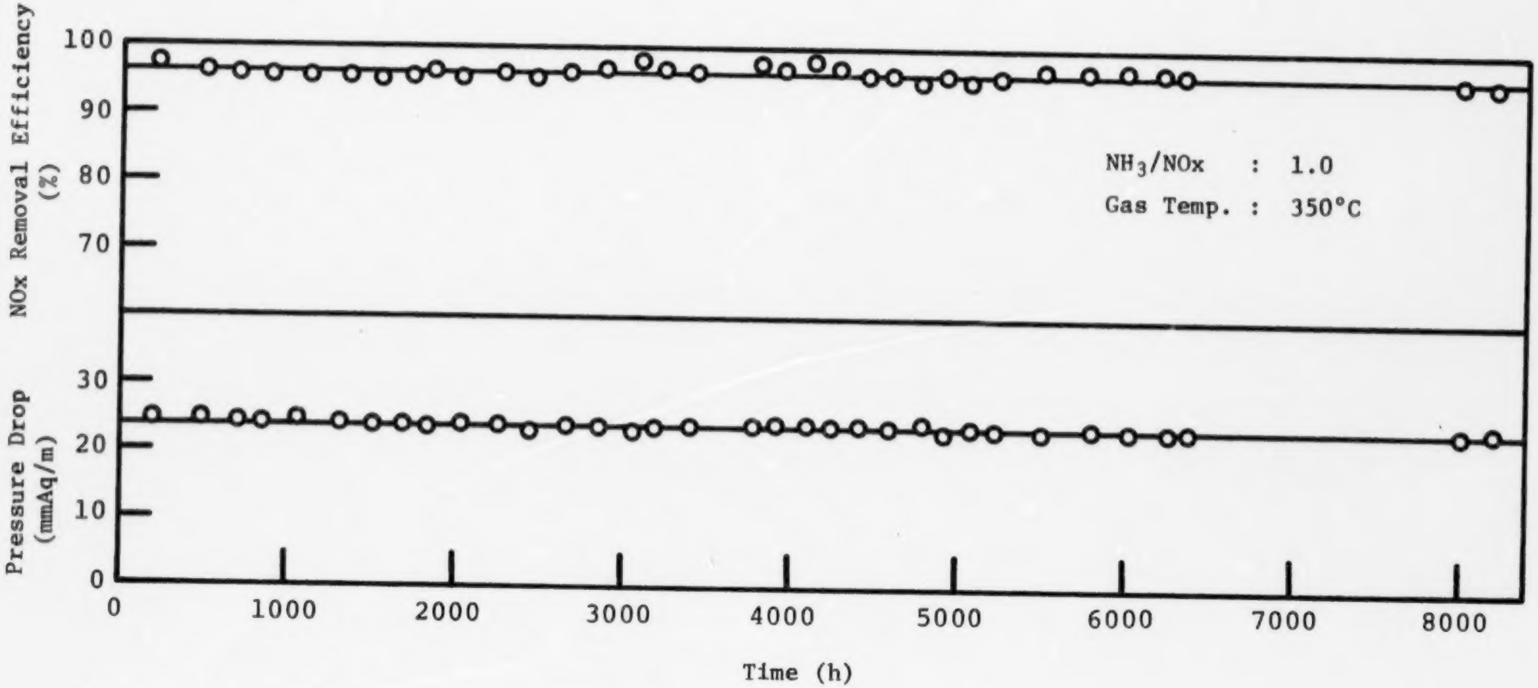


Figure 9. Durability Test of Pipe-Shaped Catalyst for the Economizer Outlet Gas of the Coal-Fired Boiler

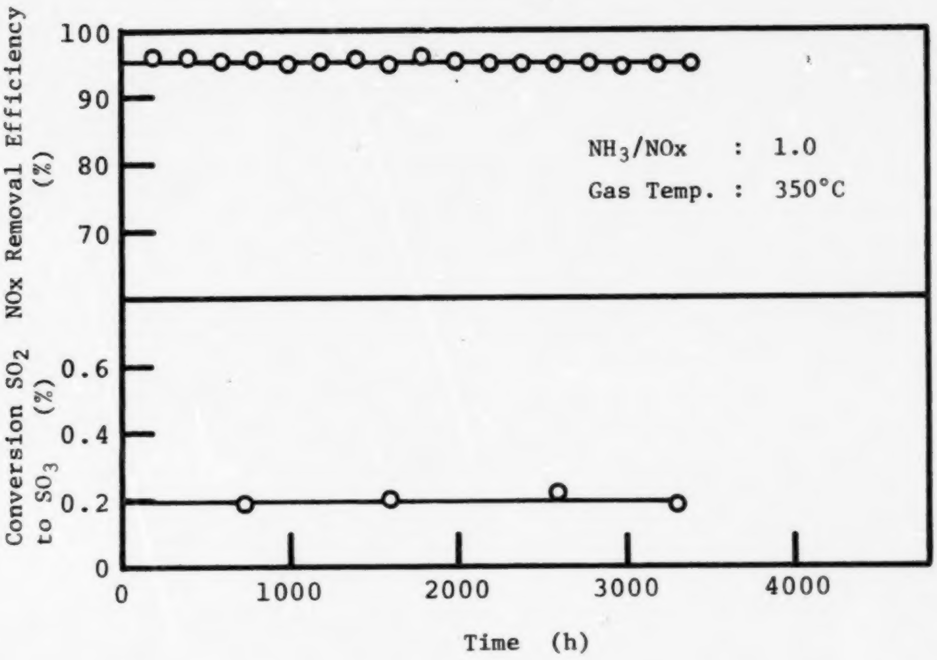
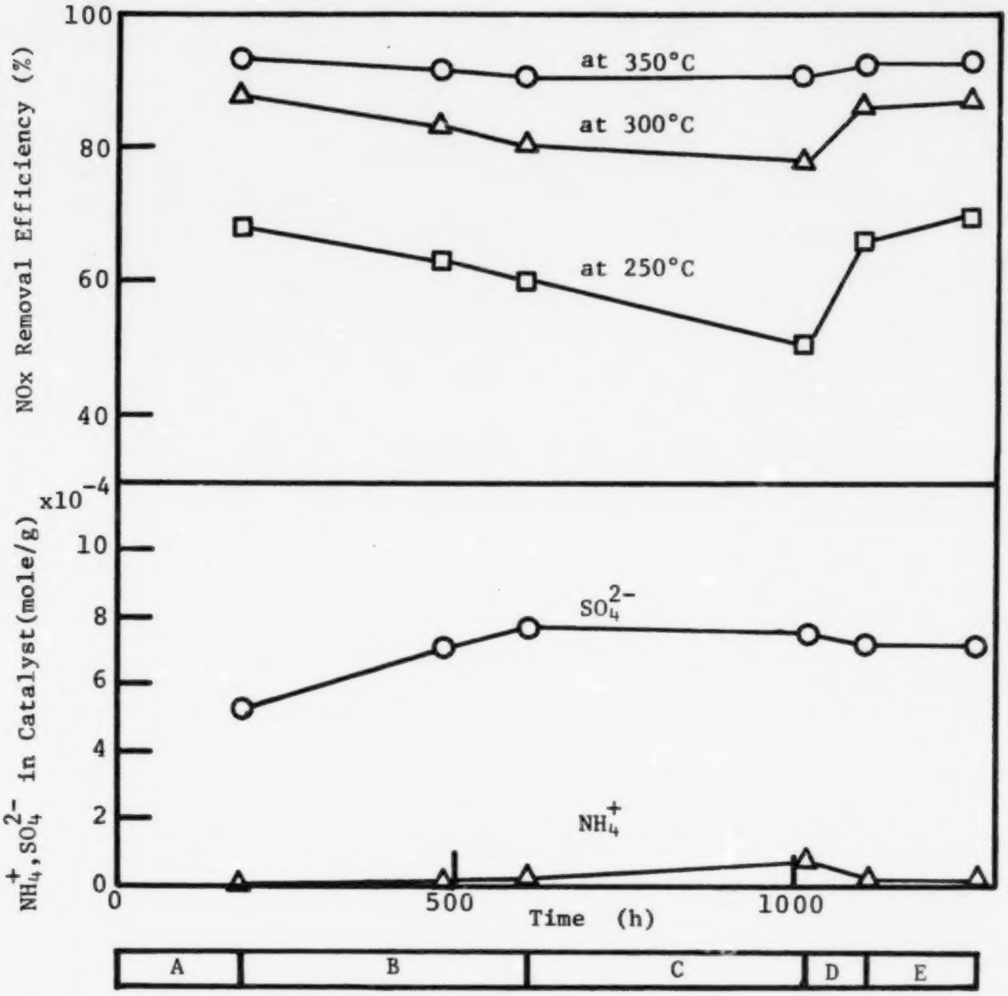
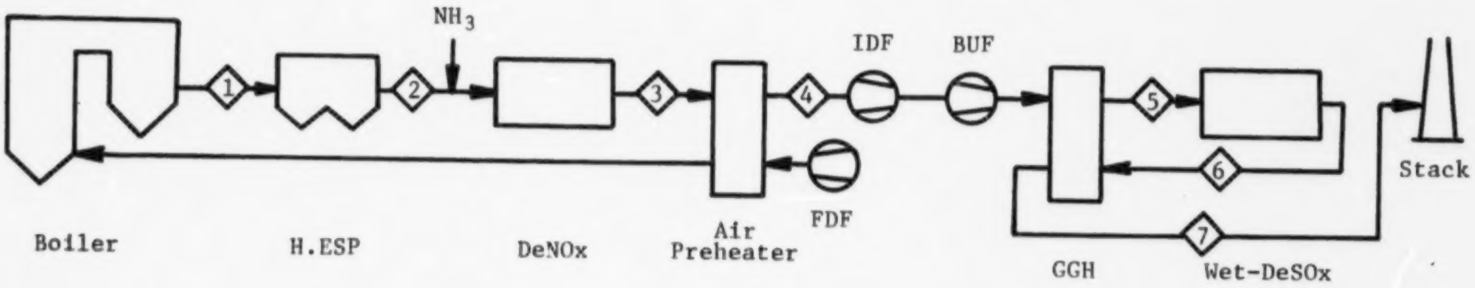


Figure 10. Durability Test of Improved Catalyst for the Economizer Outlet Gas of the Coal-Fired Boiler



A : Operation at 350°C
B : Cyclic Operation at 300°C and 270°C
Without NH_3 at 270°C
C : Cyclic Operation at 300°C and 270°C
D : Operation at 350°C
E : Cyclic Operation of Start-up and Shut-down

Figure 11. Change of Catalitic Activity and NH_4^+, SO_4^{2-} in Catalyst with Low Load Operation and Start-up/Shut-down Operation


NH3
IDF
BUF
1
2
3
4
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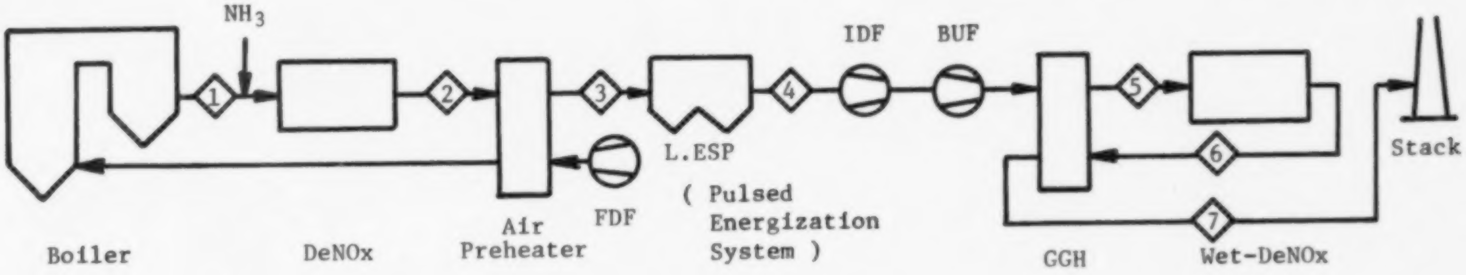
6
7
Stack
Boiler
H.ESP
DeNOx
Air Preheater
FDF
GGH
Wet-DeSOx

		①	②	③	④	⑤	⑥	⑦
Gas Volume	Nm^3/h (Wet Basis)	1700000	1714000	1723600	1861500	1913600	1967400	1915300
Gas Temperature	°C	380	380	380	150	86	45	106
Conc. of Fly Ash	mg/Nm^3 (Dry Basis)	20000	30	30	30	30	20	20
Conc. of SOx	ppm (Dry Basis)	1000	990	985	910	889	50	50
Conc. of NOx	ppm (Dry Basis)	300	300	30	30	30	30	30
Conc. of NH_3	ppm (Dry Basis)			5	5*	5*	4*	4*
Conc. of SO_3	ppm (Dry Basis)	7	7	15	5* 10	5* 10	4* 8	4* 8

* : Reduced Value of NH_4HSO_4 to NH_3 or SO_3

Figure 12. Flow Diagram of Total Flue Gas Treatment Process and Example of Material Balance for a 500 MW Coal-Fired Boiler (Low Dust DeNOx Process)



NH3
IDF
BUF
1
2
3
4
5
6
7
L.ESP
(Pulsed Energization System)
FDF
Air Preheater
Boiler
DeNOx
GGH
Wet-DeNOx
Stack

		①	②	③	④	⑤	⑥	⑦
Gas Volume	Nm^3/h (Wet Basis)	1700000	1709600	1846400	1849700	1901500	1953600	1901800
Gas Temperature	°C	380	380	150	150	86	45	106
Conc. of Fly Ash	mg/Nm^3 (Dry Basis)	20000	19900	18400	10	10	10	10
Conc. of SOx	ppm (Dry Basis)	1000	990	920	920	895	50	50
Conc. of NOx	ppm (Dry Basis)	300	300	30	30	30	30	30
Conc. of NH_3	ppm (Dry Basis)		5	5^*	Trace	Trace	Trace	Trace
Conc. of SO_3	ppm (Dry Basis)	7	15	5^* 10	Trace	Trace	Trace	Trace

* : Reduced Value of NH_4HSO_4 to NH_3 or SO_3

Figure 13. Flow Diagram of Total Flue Gas Treatment Process and Example of Material Balance for a 500 MW Coal-Fired Boiler (High Dust DeNOx Process)

TREATING FLUE GAS FROM COAL-FIRED BOILERS FOR NO_x REDUCTION WITH THE SHELL FLUE GAS TREATING PROCESS

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ABSTRACT

Copper as copper sulfate ($CuSO_4$) is one of the group of metals which at tempeatures of 350-450°C catalyzes the selective reduction of NO_x in flue gas to nitrogen and water with ammonia (NH_3). Conversions and efficiency (ammonia utilization) are high, resulting in low concentrations of both NO and NH_3 in the treated gas.

If flue gas containing both sulfur and nitrogen oxides and the reductant ammonia is processed over copper at 400°C, the copper is converted first to the oxide, then to the sulfate, and NO_x reduction begins. As the conversion to copper sulfate continues, the NO_x content of the treated gas decreases to a minimum value and the SO_x increases.

Copper sulfate can be reduced with a variety of fuels, e.g., H_2, CO, CH_4, etc., at 400°C, yielding a concentrated stream of SO_2, along with water and the copper in elemental form.

Thus, the copper system provides the technical base for flue gas treating capable of SO_x reduction, NO_x reduction, and the simultaneous reduction of both. It offers the potential of a dry process, without by-products, and with modest energy requirements.

SFGT PROCESS

The application of this technology to a NO_x-only process is in the form of a fixed bed reactor with provisions for ammonia introduction and mixing. The reactor is of special design to accommodate fly ash. The catalyst is contained in woven wire baskets suspended in the gas stream in such a way that the gas flows in the open channels between baskets (see Figure 1). The reactants enter and the products leave the catalyst bed by radial diffusion. The design is modularized in units or cells, one-half meter square by one-meter long, which can be stacked one atop the other for the required space time. The flue gas from one half MW, i.e., 1,000 SCFM, is processed in a single stack of cells.

For SO_x removal, the operation is cyclic and requires that the reactor be isolated from the flue gas circuit for regeneration, producing a concentrate of SO_2 and restoring the copper to elemental form. Continuous flue gas processing is achieved with multiple reactors, at least one of which is always in regeneration. A unit designed for SO_x removal can operate with NO_x reduction only by elimination of the regeneration step, and as a simultaneous NO_x/SO_x unit by addition of ammonia.

This reduction practice is called the SFGT process. Commercial applications have been in operation since 1973 treating flue gas from various fuels not including coal.

PILOT/DEMONSTRATION UNIT

UOP has operated an SFGT pilot plant for several years at the Tampa Electric Company's Big Bend Station near Ruskin, Florida. The pilot unit treats a slip stream of flue gas from one of the station's coal-fired boilers as the gas leaves the economizer. A simplified process flow is shown in Figure 2. De-SO_x only, simultaneous de-SO_x/de-NO_x and, most recently, de-NO_x-only operations have been conducted. The current program is sponsored by the U.S. Environmental Protection Agency.

The demonstration has involved two acceptor/catalysts: the first is in commercial application, the second, a prototype, is a more active formulation. The standard experimental procedure for each has been to establish a reference performance under fixed operating conditions using air plus pollutants followed by an evaluation on flue gas. Reference performance is again obtained with air plus pollutants before completion of the run. These tests are supported by similar evaluations in a laboratory unit.

SUMMARY OF DEMONSTRATION PROGRAM

NO_x-only evaluation of the first acceptor/catalyst was made on a fully sulfated acceptor. For four weeks, 90% NO_x reduction was achieved with a 1.25 ratio of ammonia to NO_x. The effect of the ratio on the de-NO_x performance in the reference test with air is shown in Figure 3; with flue gas, the result is the same but for a translation of the curve requiring an increase in the ratio of 10%. It has been characteristic of this system in this service that introduction of flue gas is followed by an immediate drop in performance and then stable operation.

Another characteristic of the system is that space time has little effect on NO_x reduction, that is, an increase of gas rate by 50% is accompanied by a similar percent increase in the de-NO_x reaction rate resulting in the same NO_x reduction. Both these characteristics suggest that the reactor is limited by the depletion of a reacting component which in this case is expected to be ammonia.

In the simultaneous NO_x/SO_x testing with the first acceptor/catalyst, the unit was operated for a period of approximately four weeks with 90% SO_x reduction and 70% NO_x reduction with a 1.25 ratio of ammonia to NO_x. As in the NO_x-only tests, introduction of flue gas into a clean reactor system results in an immediate drop in performance followed by stable operation. The change in performance is due to a partial blinding of the screens and the filling of the interstitial volume with fly ash.

Reference performance for the more active acceptor/catalyst with air plus pollutants is shown in the on-line analyzer strip charts in Figure 4. Time is right-to-left and was arbitrarily chosen at 99 minutes. Half-way through the acceptance, SO_x/NO_x instantaneous reduction is 98/91%; cumulative reduction at 99 minutes is 96.0/89.7.

In Figure 5 is shown the same strip charts when processing flue gas at the same conditions. The corresponding values of SO_x/NO_x reduction are 90.6/92.5 and 88.9/85.2. Note that de-SO_x performance shows the usual decline with high loadings of fly ash and SO_2 content (loading of copper with sulfur), but the instantaneous de-NO_x performance remains high and the cumulative reduction is decreased due to the "slip" of NO in the first ten minutes of the cycle while the copper is converted to copper oxide.

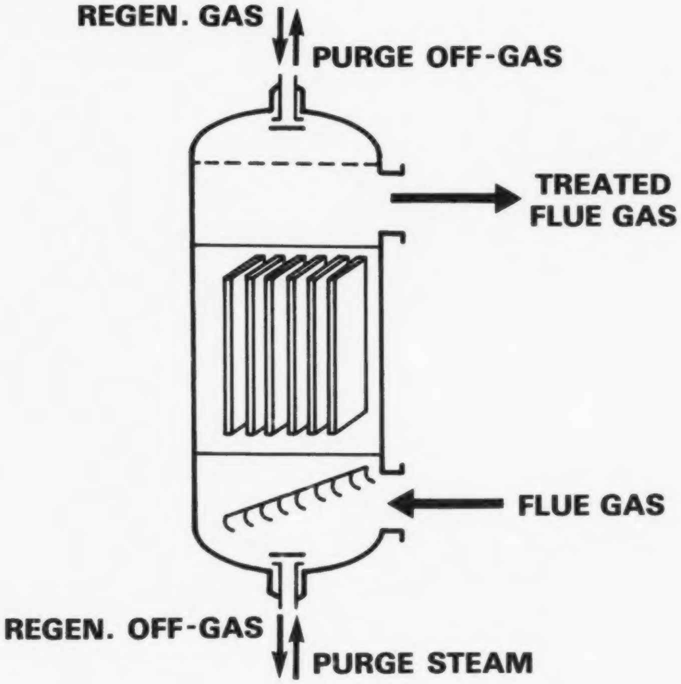
In Figure 6, also with flue gas, the rate of formation of $CuSO_4$ has been reduced to achieve 90% de-SO_x in 99 minutes, and the initial slip of NO_x decreased by delaying the injection of NH_3 for a few minutes into acceptance and with a preoxidation of the copper following regeneration. After 99 minutes, the cumulative reduction of SO_x/NO_x was 90.9 and 90.0.

The acceptor/catalyst has proven to be quite stable in this service. During stability tests carried out in 1974-1976, there was 13,000 cycles of oxidation-reduction with no significant deterioration in performance. This stability has been further supported in the current demonstration work, both in the field and the laboratory.

Operation of the TECO unit will be concluded with the testing of a de-NO_x catalyst of high activity that can function over the temperature range of 200-450°C. This catalyst is at present in commercial service at 200°C but not on coal. The performance will be determined in the parallel-passage reactor over a range of temperatures, space velocities, and NH_3/NO_x ratios with coal-derived flue gas.

FIGURE 1

THE PARALLEL PASSAGE REACTOR


REGEN. GAS
PURGE OFF-GAS
TREATED FLUE GAS
FLUE GAS
REGEN. OFF-GAS
PURGE STEAM

UOP 163-3
UOP 576-1

FIGURE 2

SIMPLIFIED FLOW SCHEME OF SFGD DEMONSTRATION UNIT FOR COAL FIRED UTILITY BOILER AT TAMPA ELECTRIC, FLORIDA

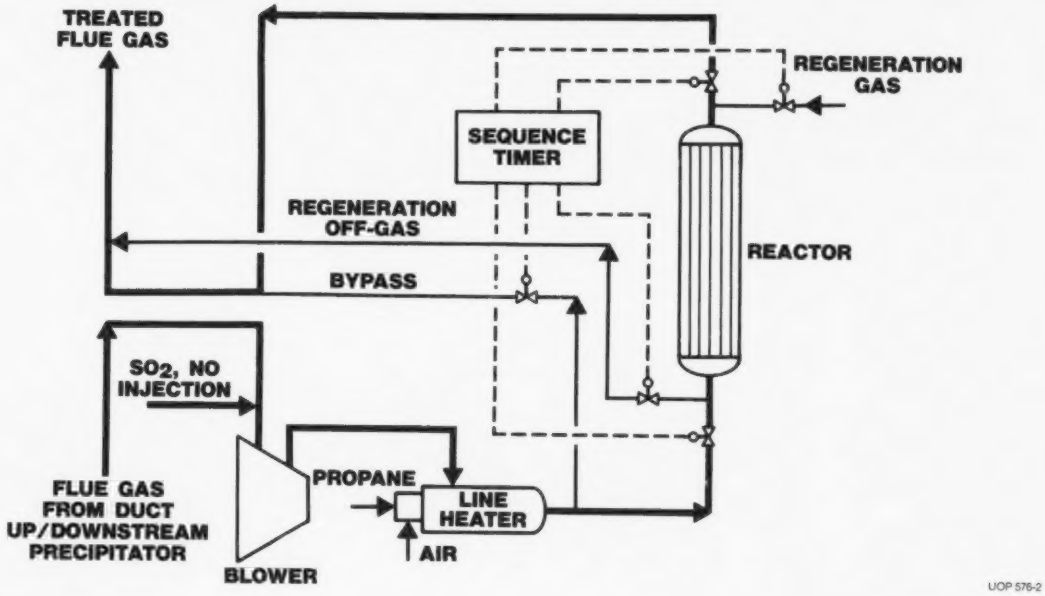
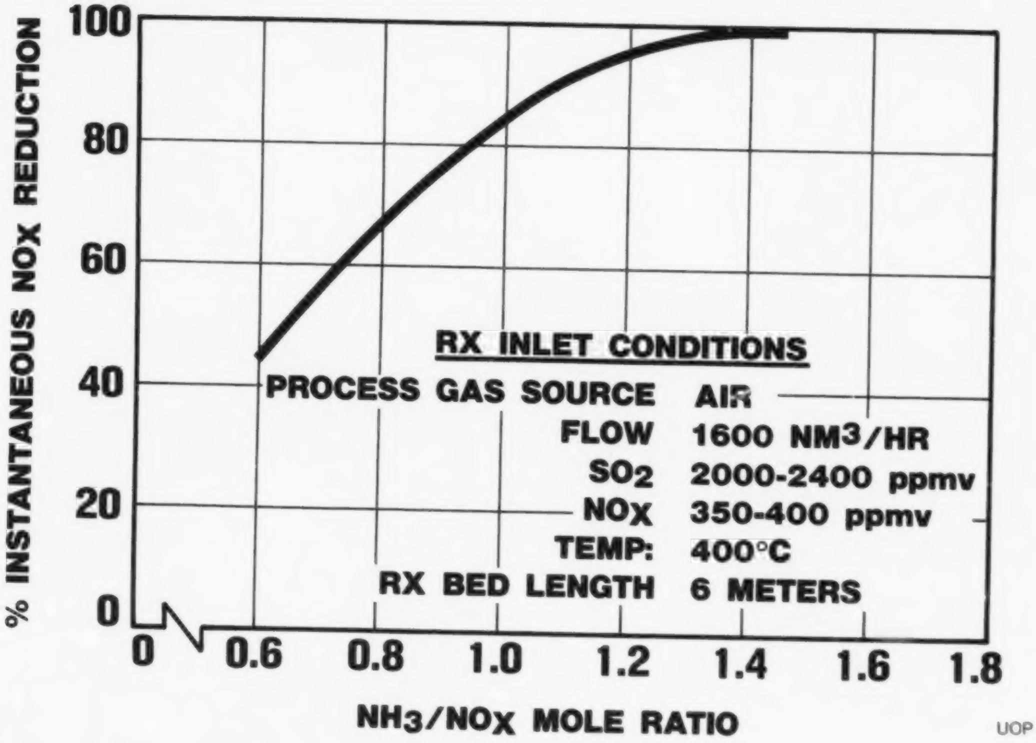

TREATED FLUE GAS
REGENERATION GAS
SEQUENCE TIMER
REGENERATION OFF-GAS
REACTOR
BYPASS
SO_2, NO INJECTION
FLUE GAS FROM DUCT UP/DOWNSTREAM PRECIPITATOR
PROPANE
LINE HEATER
AIR
BLOWER
UOP 576-2

FIGURE 3

NO_X REDUCTION vs. NH_3/NO_X RATIO AT TAMPA ELECTRIC DEMONSTRATION UNIT WITH A COMPLETELY SULFATED BED



RX INLET CONDITIONS

PROCESS GAS SOURCE	AIR
FLOW	1600 NM^3/HR
SO_2	2000-2400 ppmv
NO_X	350-400 ppmv
TEMP:	400°C
RX BED LENGTH	6 METERS

UOP 576-3

FIGURE 4

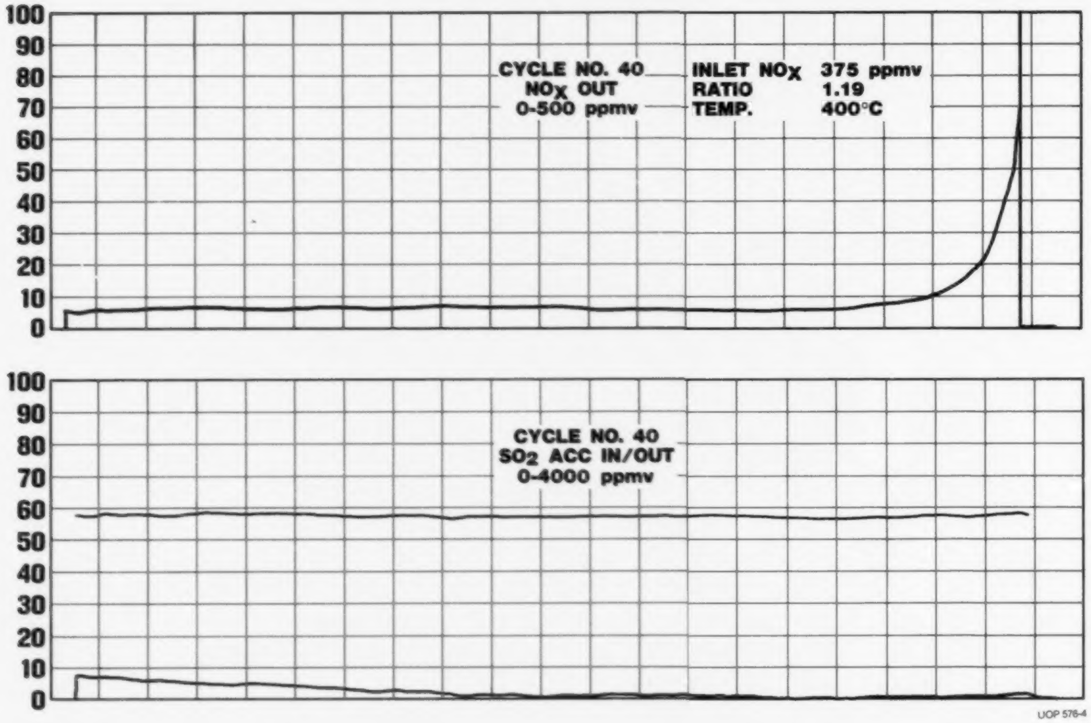

100
90
80
70
60
50
40
30
20
10
0
CYCLE NO. 40
NO$_X$ OUT
0-500 ppmv
INLET NO$_X$ 375 ppmv
RATIO 1.19
TEMP. 400°C
100
90
80
70
60
50
40
30
20
10
0
CYCLE NO. 40
SO$_2$ ACC IN/OUT
0-4000 ppmv
UOP 578-4

FIGURE 5

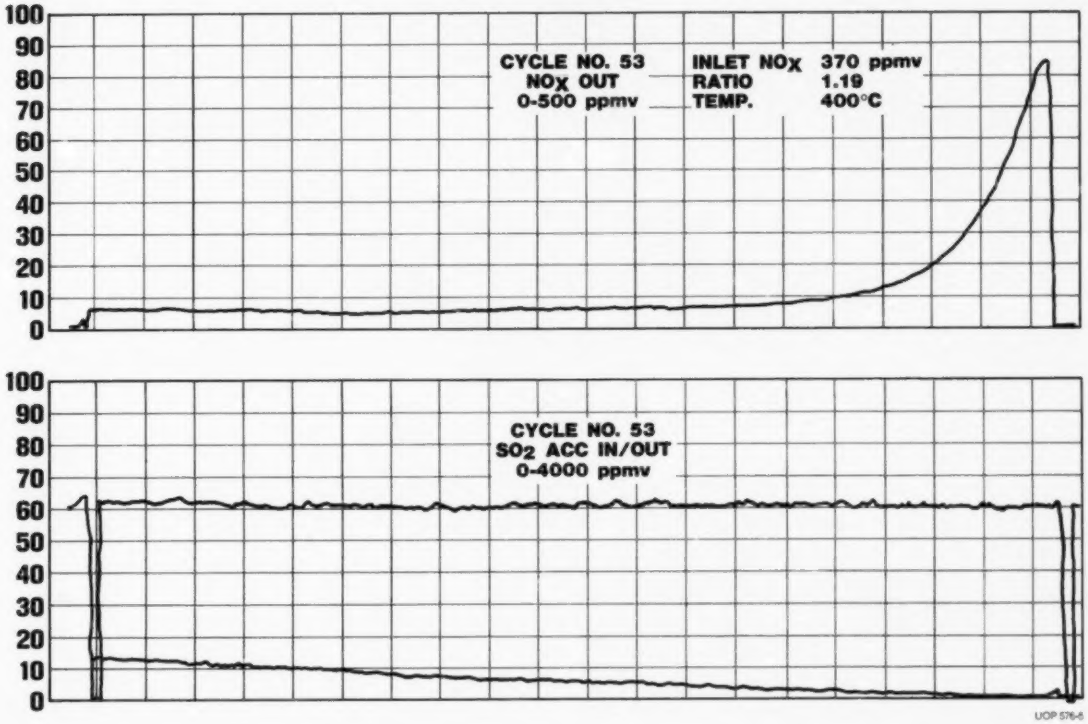
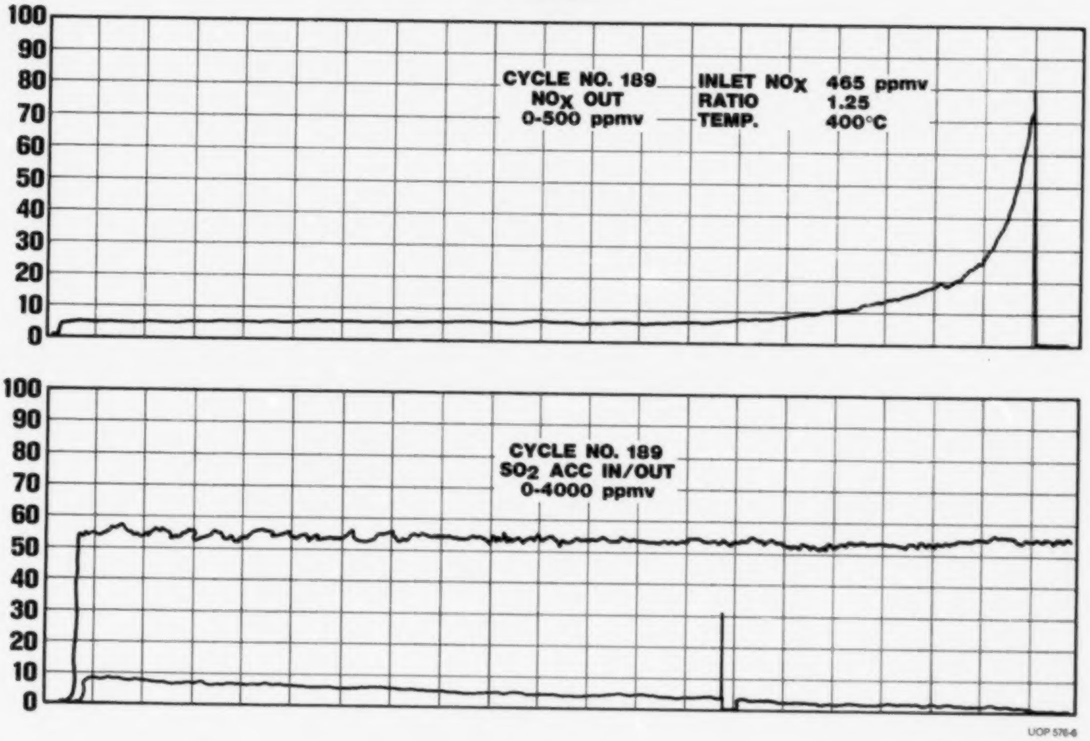

100
90
80
70
60
50
40
30
20
10
0
CYCLE NO. 53
NOX OUT
0-500 ppmv
INLET NOX 370 ppmv
RATIO 1.19
TEMP. 400°C
CYCLE NO. 53
SO2 ACC IN/OUT
0-4000 ppmv
UOP 576-6

FIGURE 6


100
90
80
70
60
50
40
30
20
10
0
CYCLE NO. 189
NO_X OUT
0-500 ppmv
INLET NO_X 465 ppmv
RATIO 1.25
TEMP. 400°C
100
90
80
70
60
50
40
30
20
10
0
CYCLE NO. 189
SO_2 ACC IN/OUT
0-4000 ppmv
UOP 576-6

THE HITACHI ZOSEN NO_x REMOVAL PROCESS APPLIED TO COAL-FIRED BOILERS

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ABSTRACT

Hitachi Zosen is a leading supplier of flue gas treatment systems for the removal of nitrogen oxides. They have nine commerical plants in operation. Early in 1978 Chemico Air Pollution Control Corp. whollyowned subsidiary of Envirotech Corporation acquired the North American license for the Hitachi Zosen technology, a dry process with selective catalytic reduction using ammonia. Because many sources of flue gas which require nitrogen oxide removal also contain a high level of dust, Hitachi Zosen has expended a considerable effort in developing a catalyst bed which can operate without plugging even though the gas contains particulates. Pilot plant tests using a specially designed metallic catalyst have been successfully operated on very dirty gases from steel sinter operations, and coal-fired boilers. An extensive pilot plant program has been in operation for over a year at Georgia Power Company's Plant Mitchell in Albany, Georgia. This is a 0.5 MW equivalent demonstration plant for coal-fired denitrification which is sponsored by U.S. EPA. Hitachi Zosen and Envirotech/Chemico believe that this testing has shown the effectiveness of the process over extended operating periods and the soundness of the control system and basic design.

INTRODUCTION

Hitachi Zosen began the development of selective catalysts for NO_x removal in late 1969 with basic research and laboratory testing. These were carrier-supported catalysts. By 1974, after extensive pilot plant studies, the catalysts were considered to be commercial. The Green Chemical Company, a wholly-owned subsidiary company, was then formed in June 1974 to manufacture these catalysts and started operation in 1975.

In early 1974 a contract was signed between the Idemitsu Kosan Company, LTD, one of the world's leading petroleum refineries in Japan, and Hitachi Zosen for the construction of an NO_x removal plant with a capacity of treating 350,000 NM^3/H (218,000 SCFM) of flue gases. The plant was the first of its kind and size in the world. Construction work commenced at the Chiba Refinery in May, 1975 with test operation in November the same year. NO_x removal efficiencies of 95% were demonstrated.

Following the successful start-up of the Idemitsu-Kosan system, other plants soon went into operation including a 274,000 SCFM unit at a petrochemical plant, and two plants at steel manufacturing facilities. All of these have operated successfully. (See Table 1 for a listing of commercial plants built by Hitachi Zosen).

A particularly effective physical design of the catalyst structure has been developed by Hitachi Zosen. This structure is of a metallic corrugated shape. Figure 1 illustrates this design. The key features of the catalyst are:

(1) All-alloy thin metal plate or wire mesh corrugated structure.

(2) Due to substantially reduced pressure drop across the catalyst layer, operating power costs are much lower than with conventional catalysts.

(3) A straight gas flow path prevents dust clogging.

(4) It is applicable for gases with high SO_x concentrations and high dust loadings.

One of the primary applications for Hitachi-Zosen's corrugated NOXNON catalyst is the treatment of high temperature gases with high SO_x and high dust concentration such as coal fired boiler flue gases. Due to the catalyst's non-clogging feature, the NO_x removal system reactor can be installed directly behind the economizer. Thus, expensive flue gas pretreatment for dust removal is not required.

Development work by Hitachi Zosen has been conducted at various bench-scale and prototype pilot plants to ensure the successful application of NO_x removal process technology for the treatment of dirty gases exhausted from power plants (coal-fired and high sulfur-containing heavy oil or residual oil fired), iron ore sintering plants, cement kilns, and other similar sources.

One of the most important of the pilot plant operations continues at the Electric Power Development Corporation (EPDC) power plant at the Isogo Station in Yokohama, Japan. This is a collaborative effort between EPDC and Hitachi Zosen under subsidy of the Japanese government. The excellent cooperation and contributions of the personnel of EPDC have resulted in the collection of a great deal of valuable data. Important developments, particularly in methods of gas distribution, have been achieved. There are three reactors installed, each treating about 200 NM^3/Hr of gas, plus several smaller reactors for abrasion testing. These reactors have operated essentially continually for several years.

NOXNON 600 catalyst was tested at this facility and some of the runs were over 6000 hours and were halted only because of the shutdown of the boiler for scheduled maintenance. As an example, one test was run for about 6300 hours at the end of which the targeted removal of 80% was still being attained. Ammonia slippage was very low as was the conversion of SO_2 to SO_3. These tests have established the effectiveness of NOXNON 600 catalyst, its resistance to abrasion, its long-term reactivity, and the low tendency to convert SO_2 to SO_3.

In addition to the work at EPDC, the pilot plant test facility for EPA, located in Albany, Georgia, has been operated for over a year. The results to date of this test program form the basis for this paper.

PROCESS DESCRIPTION

The Hitachi Zosen NO_x Removal Process is a selective catalytic reduction process in which nitrogen oxides in the boiler flue gas are reduced using ammonia as the reducing agent. The reduction reaction occurs in the presence of a proprietary catalyst.

Flue gas leaves the boiler economizer (Figure 2) at a temperature of 665°-745° (normal temperature range), depending on the boiler load, and is diverted from the air preheater by means of dampers into the ductwork leading to the reactor.

Ammonia vapor, diluted with air or steam, is injected into the flue gas in this ductwork in approximately equimolar ratio to the desired NO_x removal requirements. A pictorial representation of the ammonia injection system is shown in Figure 3. Careful attention is given to the design of the ammonia injection system to produce uniform mixing throughout the bulk of the flue gas.

The flue gas, containing the ammonia, enters the top of the NO_x reactor where it passes through a grid system which insures uniform gas distribution through the fixed catalyst bed, located below the grids. No dust removal is necessary prior to catalyst contact because of the non clogging design of the catalyst geometry. Nitrogen oxides are reduced by ammonia to ordinary nitrogen and water vapor over the catalyst surface. The treated gas then resumes the flow through the present boiler exit train, i.e., air preheater, and stack. Soot blowers are provided in the reactor for high dust laden gases to periodically clean the catalyst bed of any dust accumulation (Figure 4).

CATALYST CHARACTERISTICS

The gases to be treated flow in parallel to the plates of active catalyst material in Hitachi Zosen's corrugated catalysts, (see Figure 1). The catalyst bed consists of alternate rows of corrugated and flat plates. The distance between the flat plates is termed the pitch and this is typically between 6 and 16 millimeters. The pitch selection will depend upon the application and in particular the concentration of particulates in the gas to be treated. A larger pitch will have less tendency to retain accumulations of particulates but the larger pitch increases the required volume of catalyst.

CATALYST PREPARATION

The catalyst plates are arranged in a steel frame box supported by retainers. A standard module is 1.0 meter long, 1.0 meter wide, and 0.5 meter deep.

The activation of the catalyst follows after the corrugated catalyst assembly is made. In the NOXNON 500 catalyst, thin stainless steel plates are used. The surface is first converted to an aluminum alloy which is then treated with an aluminum dissolving solution rendering the surface layer porous. The steel surface may then be immersed in a solution containing active components which adhere to the porous surface layer. A permanent bond is obtained by further proprietary treatment.

A newer development is NOXNON 600 catalyst which, instead of plates, uses a metallic mesh material which is then coated with a porous carrier in which the active components are supported. The NOXNON 600 is considerably lighter in weight and contains more active material for a given volume of catalyst.

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The active components of the catalyst consist of vanadium and titanium compounds. Other components are added to increase resistance to fly ash abrasion.

CATALYST ACTIVITY MEASURE

Catalyst activity is calculated by the following equation and is independent of NO_x concentration, and SO_2 or SO_3 concentrations.

$$-\ln(1-X) = K(A/F)$$

where X = fraction removal of NO_x
A = surface area of catalyst
F = flue gas volumetric flow rate
K = apparent reaction rate coefficient

MEASURE OF GAS VELOCITY

The concept of space velocity is normally applied to catalysts which are granular, cylindrical, ring, extrudate, and so on. However, the concept of space velocity is not useful in designing for the use of corrugated catalysts because of this unique structure. Instead of space velocity, Hitachi Zosen uses area velocity (A.V.), which is defined as flue gas volume flow rate per unit of apparent catalyst surface:

$$(M^3/M^2\text{-Hr.})$$

The particular A.V. used in the Hitachi Zosen design depends upon the percent removal of NO_x required, the gas temperature, and other factors. However, the velocities are relatively high.

ABRASION

The catalyst can be abraded by the action of fly ash. Over a period of time the continuous abrasive blasting of the catalyst surface by the high velocity, dust laden flue gas will wear away the active catalyst material. The degree of abrasion or ash-cut has been studied by Hitachi Zosen and is estimated by the following equation:

$$A = a\,(c)\,(v^2)\,(t)$$

where A = decrease of catalytic activated layer by ash-cut

a = constant

c = concentration of fly ash

v = superficial linear velocity of gas across catalyst surface

t = elapsed operating time

A great deal of effort has gone into increasing the time that the catalyst can be used in contact with particulate laden flue gas. Special additives have been developed which are incorporated into the catalyst to increase its abrasive resistance. Precautions are taken to ensure good gas distribution so as to reduce areas of high gas velocity. These steps have been successful in allowing operations of a year or more at pilot operations in Japan where dust laden flue gases have been tested. Hitachi Zosen catalyst life of two years or more with typical coal-fired boiler conditions are obtainable.

CATALYST REGENERATION

The catalyst life is expected to be at least two years. Regeneration of the catalyst is not needed during the planned catalyst life period. At the end of this period the old catalyst would be scrapped and new catalyst installed.

CHEMISTRY OF PROCESS

The following reactions are postulated:

$$4\ NO + 4NH_3 + O_2 = 4\ N_2 + 6H_2O \quad (1)$$
$$6\ NO + 4NH_3 = 5\ N_2 + 6H_2O \quad (2)$$
$$2\ NO_2 + 4NH_3 + O_2 = 3\ N_2 + 6H_2O \quad (3)$$
$$6\ NO_2 + 8NH_3 = 7\ N_2 + 12H_2O \quad (4)$$
$$4\ NH_3 + 3O_2 = 2\ N_2 + 6H_2O \quad (5)$$

The first two reactions are probably those that occur within the reactor. As we have experienced only very low levels of NO_2, the reactions (3) and (4) are not of significance. Reaction (5) will only begin to occur at very high temperatures of 850°F or higher.

The reaction of NH_3 and NO is probably a combination of the equations (1) and (2). It has been found by many investigators that one mole of ammonia will react with more than one mole of NO. At the Georgia Pilot Plant it appears that the ratio is around 8:9. That is, eight moles of ammonia will react with about nine moles of NO. This ratio holds up to about 80-85% removal of NO.

The efficiency will drop off as increased removals are sought and at a 90% removal the ratio is close to 1.0.

IMPORTANT VARIABLES

The most critical variables which affect the degree of NO_x removal are the mole ratio of ammonia to NO_x, the flue gas flow rate, and the reactor temperature.

MOLE RATIO

This variable is defined as the ratio of moles of ammonia fed into the reactor to the moles of NO_x in the flue gas to be treated. Typically a ratio of 0.9 to 1.0 is required for a NO_x removal of 90% or higher. Figure 5 shows the mole ratio curve prepared from data collected at the Georgia Power pilot plant with NOXNON 600. It illustrates that the ammonia will remove somewhat more than one mole of NO_x per mole of ammonia feed up to a mole ratio of 0.7. Above this the removal becomes less efficient and the removal of more than 90% NO_x requires a lot more ammonia for the final quantities of NO_x to be reacted.

This figure also shows the slippage of ammonia which becomes appreciable at high mole ratios. At a 90% removal of NO_x the slippage is still relatively low, below 10 PPM.

FLUE GAS FLOW RATE

Increased flow rates provide decreased contact time with the catalyst and consequent lower removal efficiencies. However, at the normal range of operations the changes in removal are not too obvious at varying flow rates. Figure 7 shows results of tests at the Georgia pilot plant over a range of velocities which indicate a general tendency toward lower removals at the higher flow rates but the data is mostly scattered in a narrow band. Figure 5 shows Mole Ratio vs. Removal data for two flow rates and the results are very close.

Tests at a wider range of flows performed in Japan (Figure 8) indicate a definite drop off of efficiency at very high flow rates. Very high flow rates are not recommended because of increased pressure drops. Figure 6 shows the pressure drops found at the Georgia pilot plant over the test conditions. These flows would probably be typical of commercial operations.

TEMPERATURE

The flue gas temperature in the reactor is an important variable. The effects are shown in Figures 8, 9 and 10. Normally the reactor temperature is held above 330°C (620°F) to avoid the possibility of ammonium sulfate/bisulfate formation and the reactivity of the catalyst is very high at this level. At area velocities of about 10 (Figure 8) temperatures above 300°C have little effect on the percent NO conversion. In effect, the normal expected operating temperature ranges (above 300°C) have essentially no influence over the removal efficiency.

EFFECTS OF SULFUR OXIDES

The NOXNON 500 and 600 catalysts have been developed to be able to withstand high levels of sulfur oxides. They have been run for several thousand continuous hours of testing in contact with flue gases containing 400 to 500 PPM SO_2 with no measureable decrease in activity due to the sulfur oxides.

Data from Japan provides indication of the fact that there is no effect of SO_2 concentrations on the removal of NO_x. These tests were run with NOXNON 600 catalyst at 10 M^3/M^2-Hr., with a gas composition of 500 PPM NO_x and a mole ratio of 1.0.

SO_2 (PPM)	O_2 (%)	% NO Conversion			
		200°C	250°C	300°C	350°C
0	10	55	73	86	94
250	10	54	73	85	93

For SO_3, promoted catalyst life tests with concentrated SO_3 have been used to obtain data. Even with contact with SO_3 concentrations of 30,000 PPM over several hours the reaction rate of NOXNON 500 and 600 showed no decrease after several such exposures.

EPA FLUE GAS DENITRIFICATION DEMONSTRATION PLANT

In May of 1978 Hitachi Zosen received an order from the United States Environmental Protection Agency (E.P.A.) for a 1,700 Nm^3/h (0.5 MW equivalent) capacity demonstration plant for its coal-fired power plant flue gas denitrification process.

As part of its energy strategy, the United States Government plans to establish regulations which will require newly installed power plants to use coal as their energy source instead of heavy fuel oil or natural gas. But most U.S. coals have high SO_x (sulfur oxides) and ash concentrations, and no commercial technology has been introduced to remove NO_x (nitrogen oxides) from flue gases from power stations using these coals.

Therefore, the E.P.A. decided to construct a flue gas denitrification demonstration plant to carry out technological and economical assessments on high SO_x and dust-containing coal-fired power station flue gas.

The objective of the pilot plant program is to demonstrate Hitachi Zosen's catalytic reduction process for the treatment of flue gas with ammonia for the removal of 90% of NO_x. The program will evaluate the performance of the NOXNON 500 and 600 catalysts, which have been developed by Hitachi Zosen for flue gas of high sulfur oxide and high dust content which is exhausted from coal-fired boilers.

The original planned duration of the test program included a three-month start-up and test operation period plus three months of continuous operation. Varying levels of nitrogen oxide and sulfur oxide concentrations were tested along with varying temperatures and gas flow rates. This program has been extended and the test operation will be run for over a year.

Through the generosity and support of Georgia Power Company the pilot plant host site is the Plant Mitchell of GPC at Albany, Georgia. The flue gas is obtained from Unit #3. This unit was built in 1964 and consists of a coal-fired C-E Boiler and a Westinghouse Turbine-Generator with a capacity of 165 MW. Steam temperature is 1000^{o}F and the pressure 1800 PSI.

Flue gas for the pilot plant is obtained by connecting a 14" duct into the power plant duct after the economizer. The slipstream of flue gas is first passed through an electric heater to control the gas temperature, ammonia is injected into the gas, and the gas then passes through the reactor vessel where the nitrogen oxide is converted to nitrogen. The cleaned gas is repressurized in a blower and returned to the power plant duct. Additional quantities of nitrogen oxide and sulfur trioxide are generated, as required, to vary the concentration in the flue gas.

Construction and operation of the pilot plant was carried out by Chemico Air Pollution Control Corporation (CAPCC), Hitachi Zosen's North American licensee. Fabrication and procurement was done within the United States as far as possible.

In implementing the contract with E.P.A., Hitachi Zosen provided the basic design, proprietary catalysts, and fabrication of the NO_x, SO_3 generating unit needed for the system. CAPCC, as major subcontractor, provided the detailed engineering and design, procurement, erection and operation of the pilot plant.

The initial NOXNON 500 catalyst charge provided excellent results for about 2000 hours of operation. Based on experience in Japan the sootblower was not operated because the pressure drop remained constant. After this period of time however, there occurred a drop in removal efficiency and an increase in pressure drop. Removals decreased from the normal 90% level to the 80% level. Intensive efforts were made to check and recheck the analytical and control equipment to make certain that it was the catalyst at fault. It became obvious that the catalyst activity had definitely decreased below the acceptable level.

A spare batch of NOXNON 500, which was at the site, was installed to replace the initial batch. The replacement material was exactly the same shape and composition as the original.

Samples of the original batch were flown back to Japan for intensive investigations. The results of these indicated that there was no loss of catalyst activity. The material was as chemically sound as originally provided. It appeared that the loss of activity was due to a masking of a portion of the catalyst surface due to a buildup of fly ash within the catalyst bed. This phenomenon was not exhibited in Japan during long-run tests at a coal-fired station. Analyses were made of the fly ash from Georgia as compared to that in Japan. One possible significant difference was that the Georgia material contained a relatively high level of unburned coal or carbon. Physical tests were performed on the fly ash and it showed that the Georgia material had a greater tendency to agglomerate at high temperatures.

One of the contributing factors to the fly ash build-up on the catalyst was the design of the ductwork. When the ducts were disassembled for an inspection of the system it was found that the design allowed a large accumulation of fly ash in a horizontal run above the reactor. Some of this fly ash undoubtedly loosened and fell onto the catalyst bed, thus blocking a portion of the flow through the reactor. This probably contributed tc the lower than expected performance of the process. The ductwork was changed soon after the inspection was made and such problems were not encountered later.

The replacement NOXNON 500 also gave excellent results for some months of operation. However, again after some 2000 hours of operation it began to exhibit a loss of activity. This happened despite daily operation of the sootblower. This catalyst was taken out of the reactor, cleaned with compressed air, and was replaced. The pressure drop, which had increased at the same time the removal decreased, returned to a normal level but the activity did not recover.

The loss of activity of the second batch of catalyst was found to be due to a physical blinding of the surface of the catalyst by fly ash. The degree of fly ash accumulation was surprising. Investigations into the causes continue. One clue is the tendency to an increased pressure drop when the system is shutdown and restarted. One theory is that deposits on the elements of the flue gas heater are released when the system is cooled down. They are then carried with the flue gas during restarts, into the reactor. The extreme temperatures on the heater elements over long periods of time probably cause physical or chemical changes in the deposited fly ash which might make it more adhesive. This altered fly ash could stick to the catalyst surface. This situation was not encountered during many months of operation in Japan probably because of the difference in the heater design and the difference in the fly ash composition. If this did actually occur in the pilot plant, it would not be expected to happen in commercial systems where flue gas heaters are not required.

The third batch of catalyst was then installed. This catalyst was of Hitachi Zosen's latest design: NOXNON 600. This is made from a metallic mesh instead of from sheets of steel and is consequently much lighter in weight. Of more significance is the fact that the pitch of this catalyst was larger:

14 mm vs. 8 mm. The openings are much wider and the tendency to clog is greatly reduced.

Because of the wider pitch, the reactor had to be lengthened to accept the new catalyst which required more volume. This NOXNON 600 catalyst has been in service since April 1980 and, as of this writing at the end of July, is providing excellent results. It is planned to test this catalyst at least until October.

PROCESS DESIGN CONSIDERATIONS

AMMONIUM SULFATE/BISULFATE FORMATION

The formation of ammonium sulfate (or ammonium bisulfate) produced by the reaction between ammonia and sulfur trioxide can cause difficulties in the process. Deposits can result in a decrease of catalyst activity due to adhesion on the catalyst-activated surface and/or blockage of the catalyst layer.

There is an increased tendency towards ammonium sulfate formation at higher levels of ammonia and sulfur trioxide and at lower temperatures. The best means of prevention is to maintain a temperature so high that this compound cannot form. This is normally around $330^{\circ}C$ ($620^{\circ}F$). If the reactor temperature, for some reason, begins to drop and approaches this temperature it would be best to stop the ammonia flow until the required minimum temperature is reattained. If deposits are formed on the catalyst by a process upset the sootblower can be effective in removing these.

Deposits on downstream equipment such as air preheaters are best avoided by maintaining a low level of ammonia slippage.

AMMONIA CONTROL SYSTEM

The pilot plant at Georgia was designed to simulate as closely as possible the control system for a full-sized commercial plant. The ammonia feed system is completely automatic and requires only that the operator select the desired mole ratio. From that point on the ammonia feed will be delivered as required to satisfy the selected ratio of ammonia to incoming nitrogen oxide.

In this control system, the flue gas flow rate and the incoming nitrogen oxide concentration are measured and the signals are multiplied to provide the mass flow of NO_x entering the reactor. This quantity is then multiplied by the set mole ratio to determine the amount of ammonia required. This signal, in turn, is relayed to the ammonia flow meter to set the ammonia flow.

This system has worked exceptionally well. This ammonia control system can probably be applied to a full-scale unit along with some refinements, such as using the outlet NO_x or outlet ammonia concentration to provide a signal to fine tune the ammonia feed rate.

One potential drawback to this control system is the measurement of the flue gas flow rate. This is difficult to do on a large boiler system because usually there are no straight runs of duct in which a flow element could be installed. Most of the flue gas passes through headers without any significant ducting. If there were a straight run, either before or after the reactor, a flow element could be installed to provide a pressure drop signal as a measure of gas flow. Such a system is used at the Georgia pilot plant and might be practical even for larger boilers.

In the absence of a flue gas flow meter other signals are available. One of these is the steam flow rate to the turbine. This is a measure of the fuel burned and indirectly a measure of the combustion gases produced. Combining this signal with the inlet NO_x level will provide a fairly accurate measure of the NO_x mass loading. This control can then be fine tuned using the outlet NO_x level or the percent removal of NO_x.

REACTOR INSTRUMENTATION

Both temperature and pressure drop are required measurements for the reactor. The temperature of the flue gas has an effect on the removal efficiency. Normally, the temperature would not be controlled and the reactor would accept whatever the gas temperature would be at the economizer exit. At the Georgia pilot plant an electric heater is supplied to control the temperature because of the high heat loss from the ductwork, typical of pilot plants.

A low temperature at the reactor inlet is of concern because of the risk of ammonium bisulfate formation. At low temperatures an interlock is provided to shut down the ammonia supply by overriding the ammonia flow controller.

The pressure drop across the reactor is an important measurement. This will indicate whether there is any tendency to plugging by fly ash or by other deposits within the catalyst bed. At an increased pressure drop the sootblower is actuated to clean the catalyst.

SOOTBLOWER CONTROL SYSTEM

Soot blowers are installed to periodically clean the catalyst bed. They can use steam or air. The system can be completely automated and initiated manually by a push button. At the Georgia pilot plant the system is set up the same as would be provided for a commercial system. Upon initiation steam or air is admitted into the electric heater which raises the temperature of the gas to 650°F. The gas is released to atmosphere until the temperature is sufficiently high. When the temperature is high enough a valve opens permitting the gas to flow into the sootblower and at the same time the sootblower moves forward across the top of the reactor blasting the hot gas down through the catalyst. After one cycle, forward and back, the system automatically stops, the remaining gas is released to the atmosphere, and the system shuts off. There are interlocks provided to ensure proper gas pressure and to ensure that the sootblower moves properly. The air or steam pressure and temperature are monitored. At the Georgia pilot plant the sootblower has been run once per shift since the NOXNON 600 catalyst was installed. Both air and steam are being tested.

AMMONIA SLIPPAGE

A certain amount of ammonia can be lost from the process without reacting. This can happen particularly at high NO_x removals when there would have to be an excess of ammonia available to approach complete reactions with the NO_x. Experience has shown that slippage of ammonia is negligible below about 80% NO_x removal. At 90% removal or above the slippage can amount to 10-20 PPM or higher.

The control of ammonia slippage is important mainly because of the risk of accumulations of ammonium sulfate/bisulfate on downstream equipment. In Japan some clients have requested targets as low as 10 PPM. The need for a continuous measurement of ammonia slippage is very obvious.

At the Georgia pilot plant ammonia slippage has been essentially nil below a mole ratio of 0.9. As the ammonia feed is increased, however, the slippage rapidly increases as can be seen in Figure 5.

The pilot plant in Albany, Georgia was originally designed to provide a continuous read out of ammonia levels in the flue gas leaving the reactor. The design used a chemiluminescent analyzer with two converters. The sample of flue gas drawn through the sample line was split. Half was sent through a high temperature converter to convert all of the ammonia and NO_2 to NO. The other half of the sample stream was passed through a low temperature converter which converted only the NO_2 to NO and did not affect the ammonia. The analyzer was designed to determine the NO level in each of the two streams in the two chambers and by difference provide the ammonia concentration. In principle this system should work but the problem was to ensure that the ammonia reached the converter.

Ammonia can be easily lost from the flue gas sample before it reaches the analyzer. It can react with SO_2 or SO_3 or it can be absorbed by condensed moisture or by deposits of solid material. We have even experienced the absorption of ammonia by certain gas filters.

Despite heroic efforts at the pilot plant it was not possible to make the ammonia analytical system operable. A good sample to the analyzer could not be ensured. Eventually they went to wet sampling methods using impingers to absorb ammonia from gas samples and a Hach colorometric method to determine the concentration of ammonia in the absorbent. This system has provided the necessary data but would not be a practical system for a commercial plant.

Further efforts continue in the design and testing of a continuous ammonia slippage monitoring system. A gas conditioning system has been designed by GCA and Charlton Associates to treat a sample of flue gas at the sample point in the duct before the sample can cool down. A sample of flue gas is filtered and then passed through an absorbent to remove sulfur trioxide. The gas is either passed through a stainless steel converter to convert the ammonia to NO or is bypassed around the converter. The gas sample is then dried and sent by the sample line to the analyzer at the control rooms. By comparing the NO level of the sample passed through the converter and the sample bypassing the converter, the ammonia analysis can be determined. This system avoids the problem of the reaction of ammonia with sulfur trioxide by absorbing the sulfur trioxide and avoids the problem of the reaction with sulfur dioxide by maintaining a minimum temperature above which the reaction cannot take place.

Eventually a workable system will be evolved which can supply a continuous record of ammonia slippage.

STARTUPS AND SHUTDOWNS

Hitachi Zosen experience indicates that no special precautions are needed during starting up and shutting down of the Hitachi Zosen NO_x removal system in coal-fired units except for the avoidance of fly ash accumulations. Ammonia injection should of course be stopped whenever the temperature drops below about 300°C to prevent ammonium sulfate/bisulfate formation on the catalyst. The reactor can follow the normal temperature changes along with the boiler with no difficulties. The catalyst has been tested through hundreds of temperature cycles without deterioration. If SO_3 condenses on the catalyst during shutdowns or startups, this produces no adverse effects and the SO_3 will evaporate when the temperature is raised to normal levels. The SO_3 condensate will not corrode the catalyst or reduce its activity.

Fly ash accumulations on the catalyst can be avoided by careful design of the ductwork to ensure that proper flow distribution is maintained at most flow rates. Soot blowing is an additional measure to avoid fly ash deposits.

FIGURE 1

NOXNON 600 CATALYST CONFIGURATION

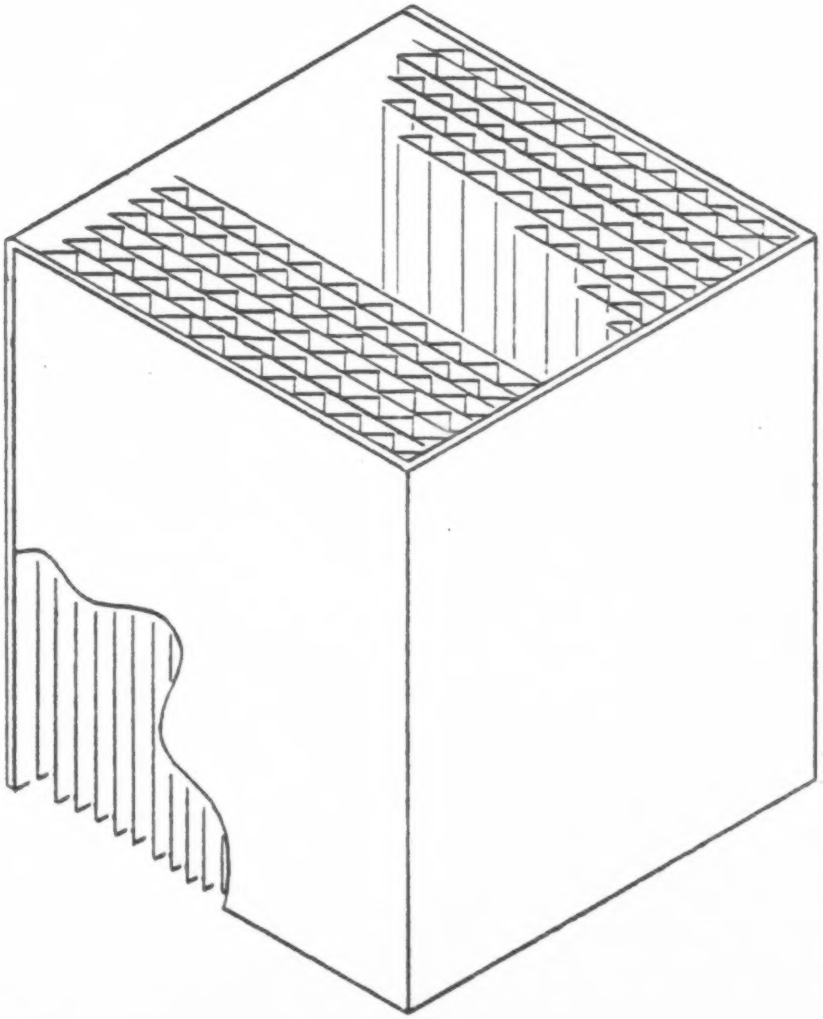


FIGURE 2

TYPICAL FLOW SHEET COMMERCIAL SYSTEM

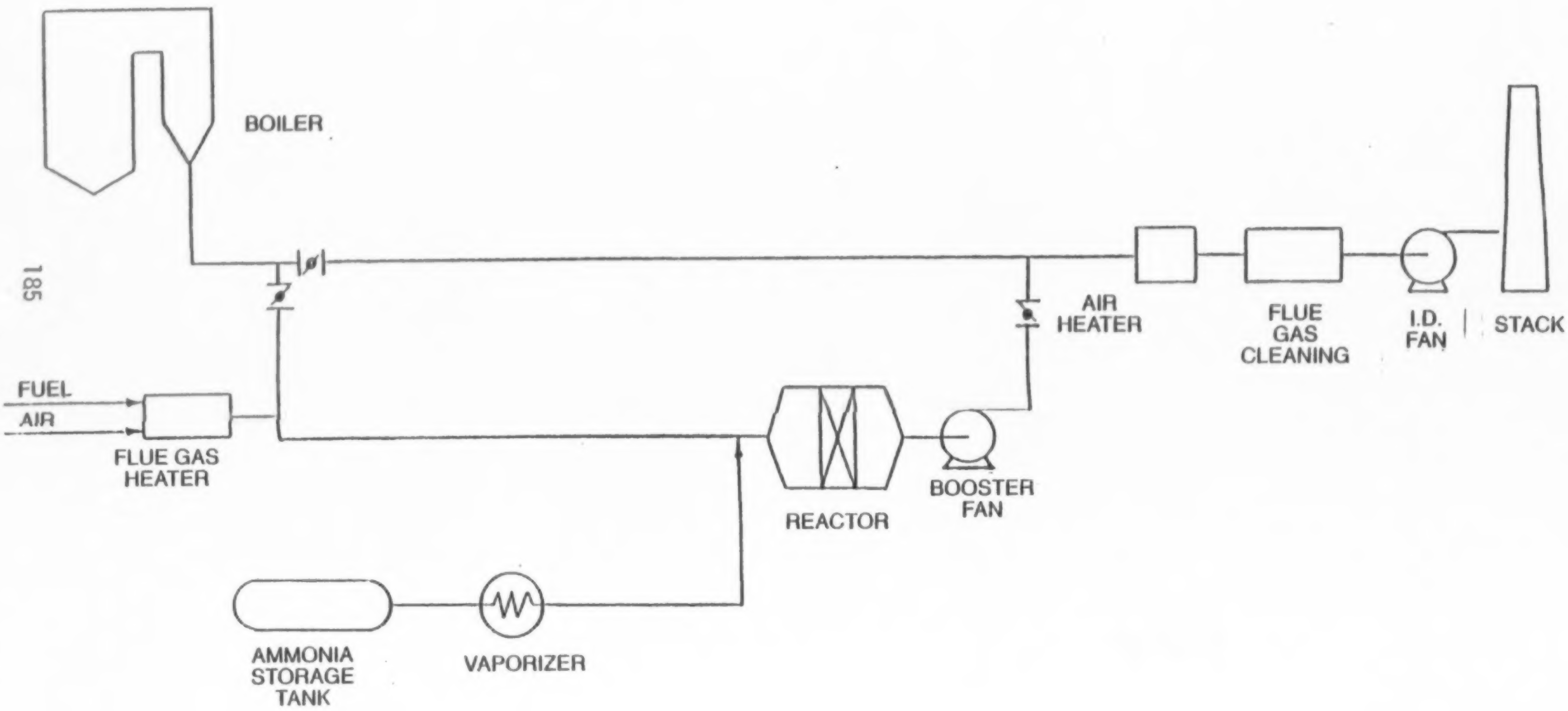
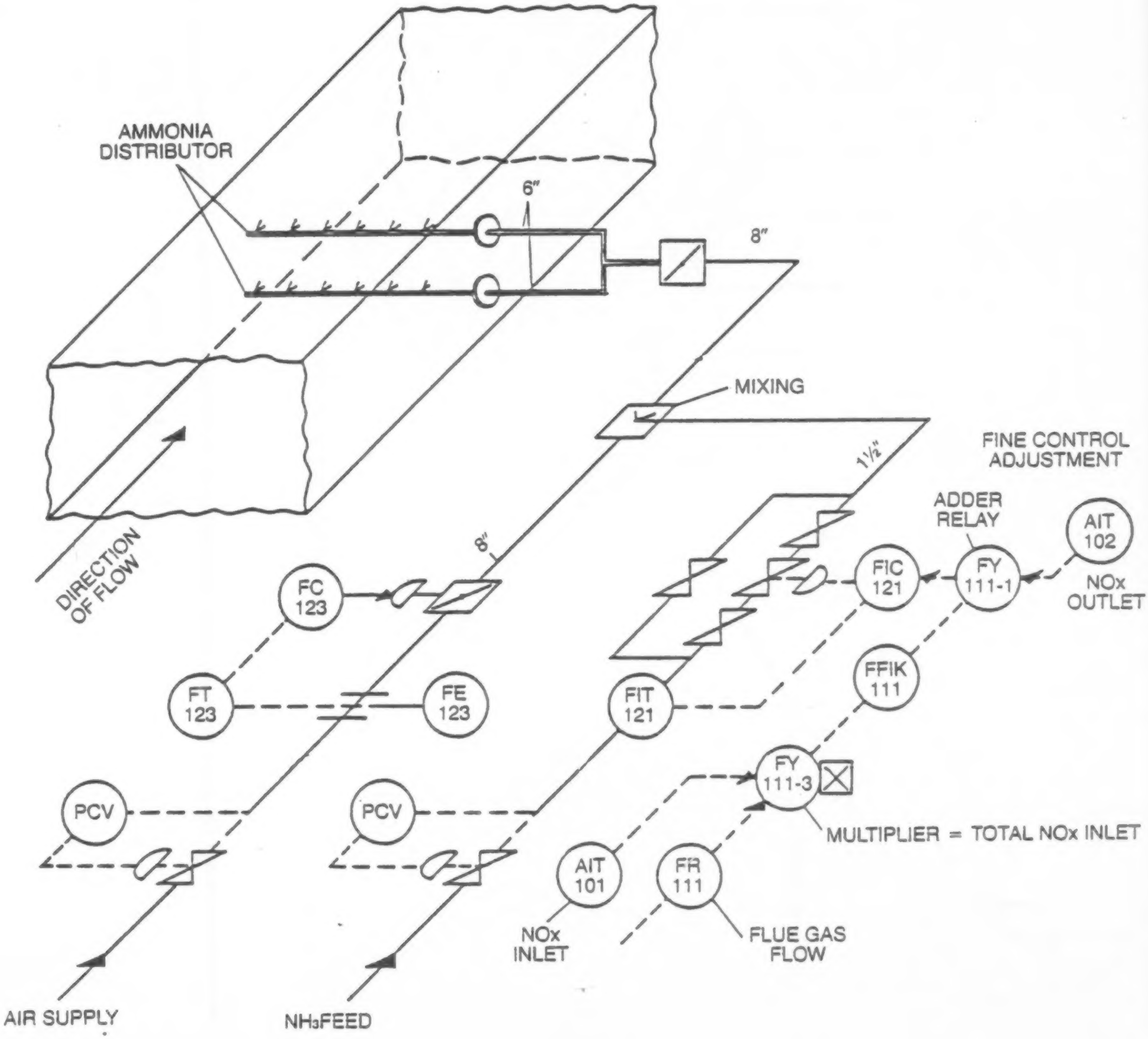

BOILER
FUEL
AIR
FLUE GAS
HEATER
AMMONIA
STORAGE
TANK
VAPORIZER
REACTOR
BOOSTER
FAN
AIR
HEATER
FLUE
GAS
CLEANING
I.D.
FAN
STACK

FIGURE 3

AMMONIA INJECTION SYSTEM


AMMONIA DISTRIBUTOR
6"
8"
MIXING
FINE CONTROL ADJUSTMENT
1½"
ADDER RELAY
AIT 102
FY 111-1
NOx OUTLET
FIC 121
DIRECTION OF FLOW
8"
FC 123
FFIK 111
FT 123
FE 123
FIT 121
FY 111-3
PCV
PCV
MULTIPLIER = TOTAL NOx INLET
AIT 101
FR 111
NOx INLET
FLUE GAS FLOW
AIR SUPPLY
NH₃FEED

186

FIGURE 4
SOOT BLOWER

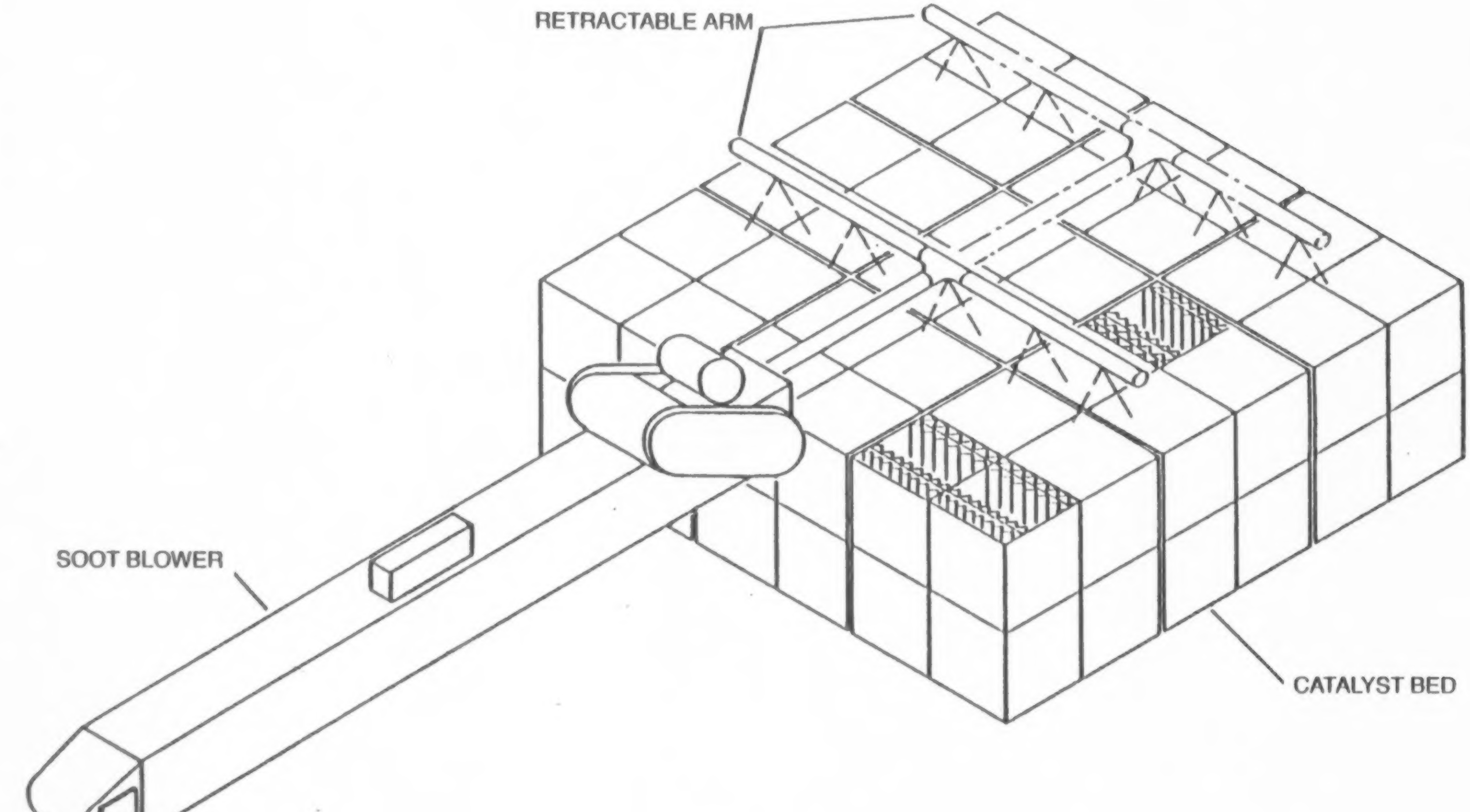
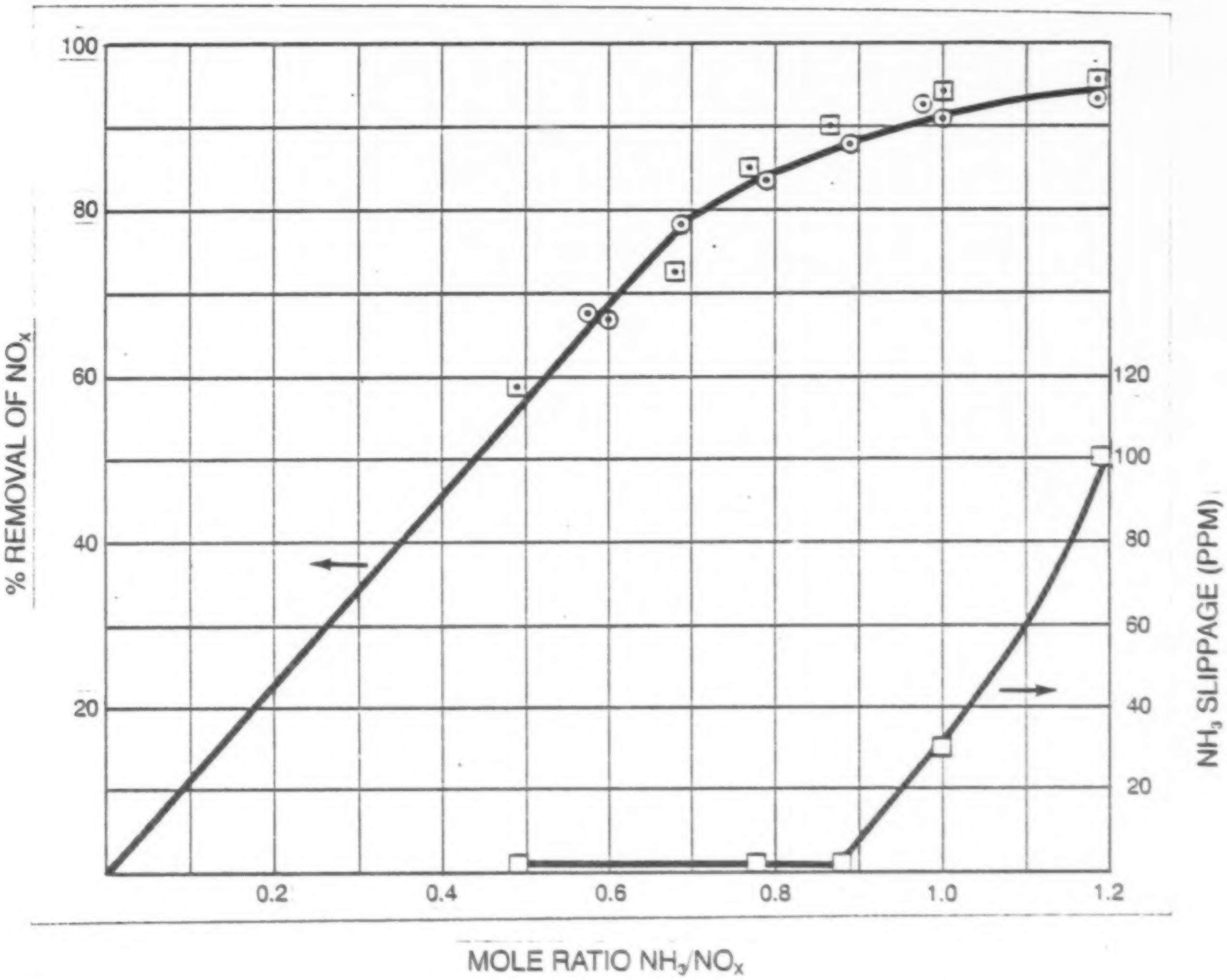

RETRACTABLE ARM
SOOT BLOWER
CATALYST BED

FIGURE 5

NO_x REMOVAL vs. MOLE RATIO



Catalyst: NOXNON 600

Temp.: 700-710°F

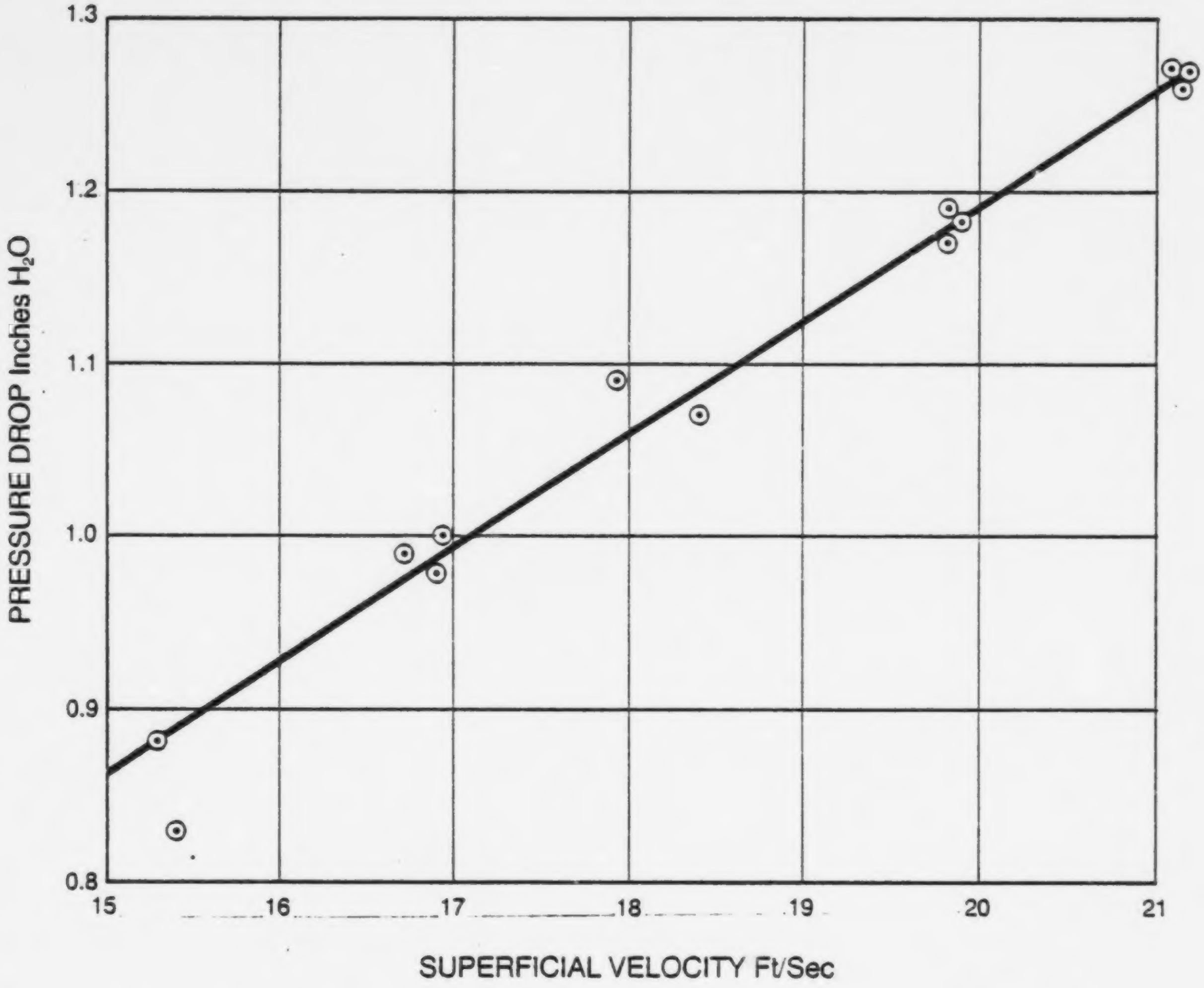
Flow Rate: ○ 1500 SCFM

□ 1300 SCFM

Inlet NO_x: 400-500 PPM

FIGURE 6

PRESSURE DROP vs. VELOCITY



NOXNON 600

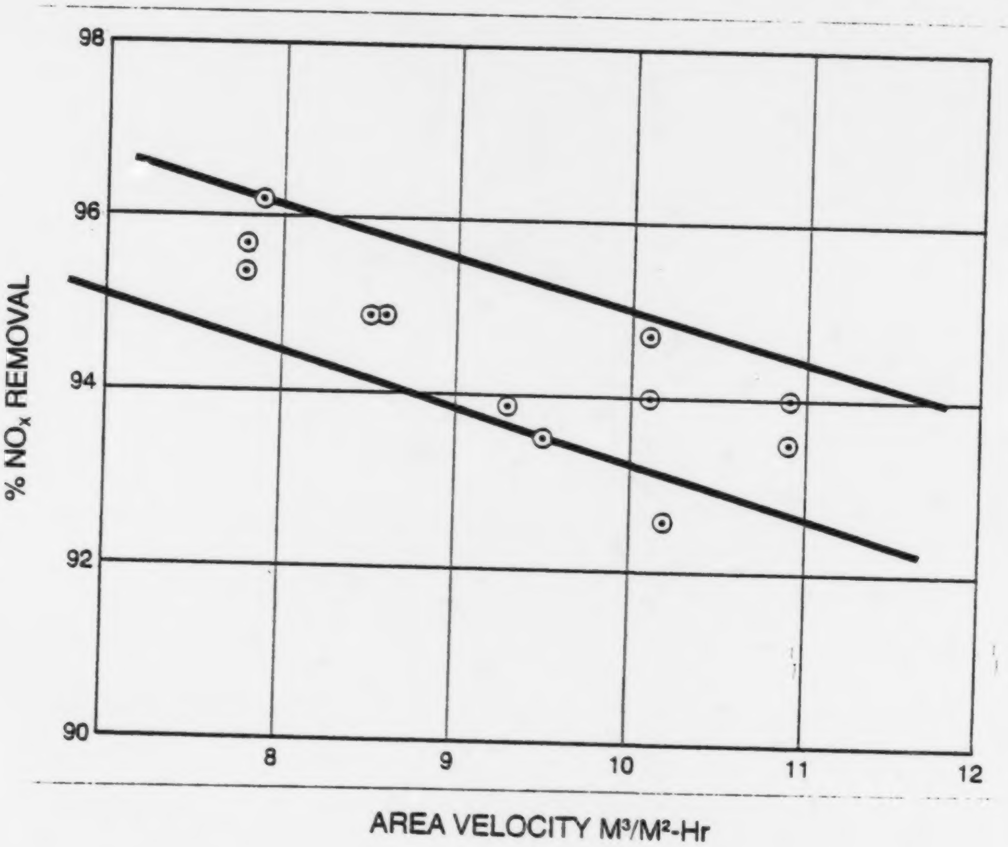
Bed Depth: 2.0 Meters

Temperature: 715°F

184

FIGURE 7

% NO_x REMOVAL vs. AREA VELOCITY



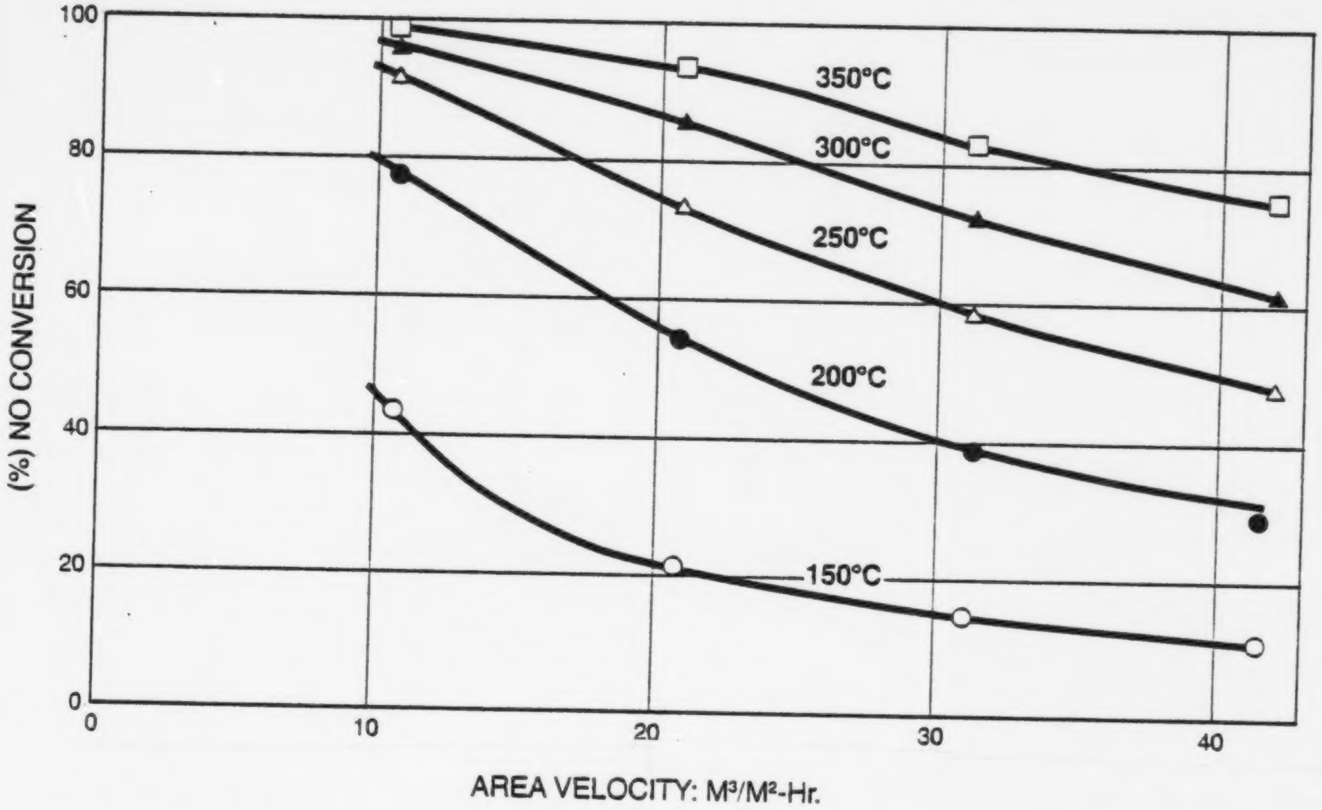
Mole Ratio: 1.0

Reactor Temp.: 720°F

Inlet NO_x Conc.: 400-450 PPM

FIGURE 8

AREA VELOCITY vs. NO CONVERSION



Catalyst: NOXNON 600

Gas Composition: 500 PPM NO

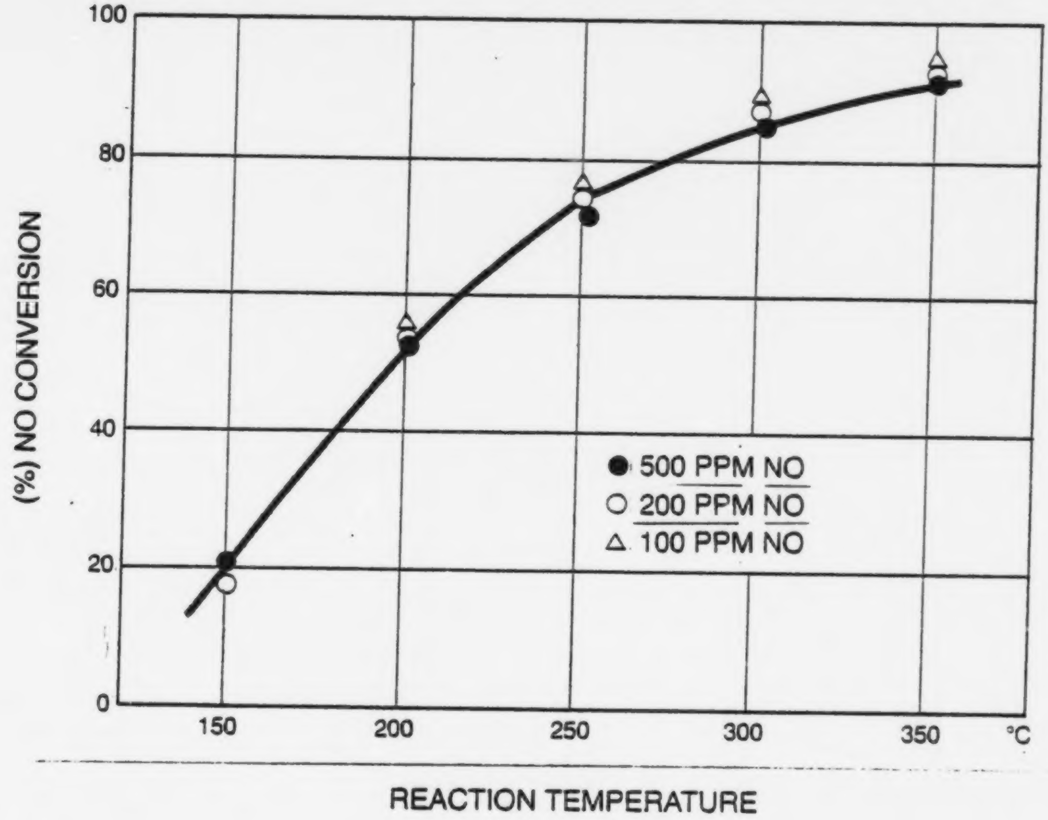
500 PPM NH_3

250 PPM SO_2

6% O_2, 10% H_2O, 10% CO_2

FIGURE 9

REACTION TEMPERATURE vs. NO CONVERSION



Catalyst: NOXNON 600

Area Velocity: 20.4 M³/M²-Hr.

Gas Composition: 250 PPM SO_2

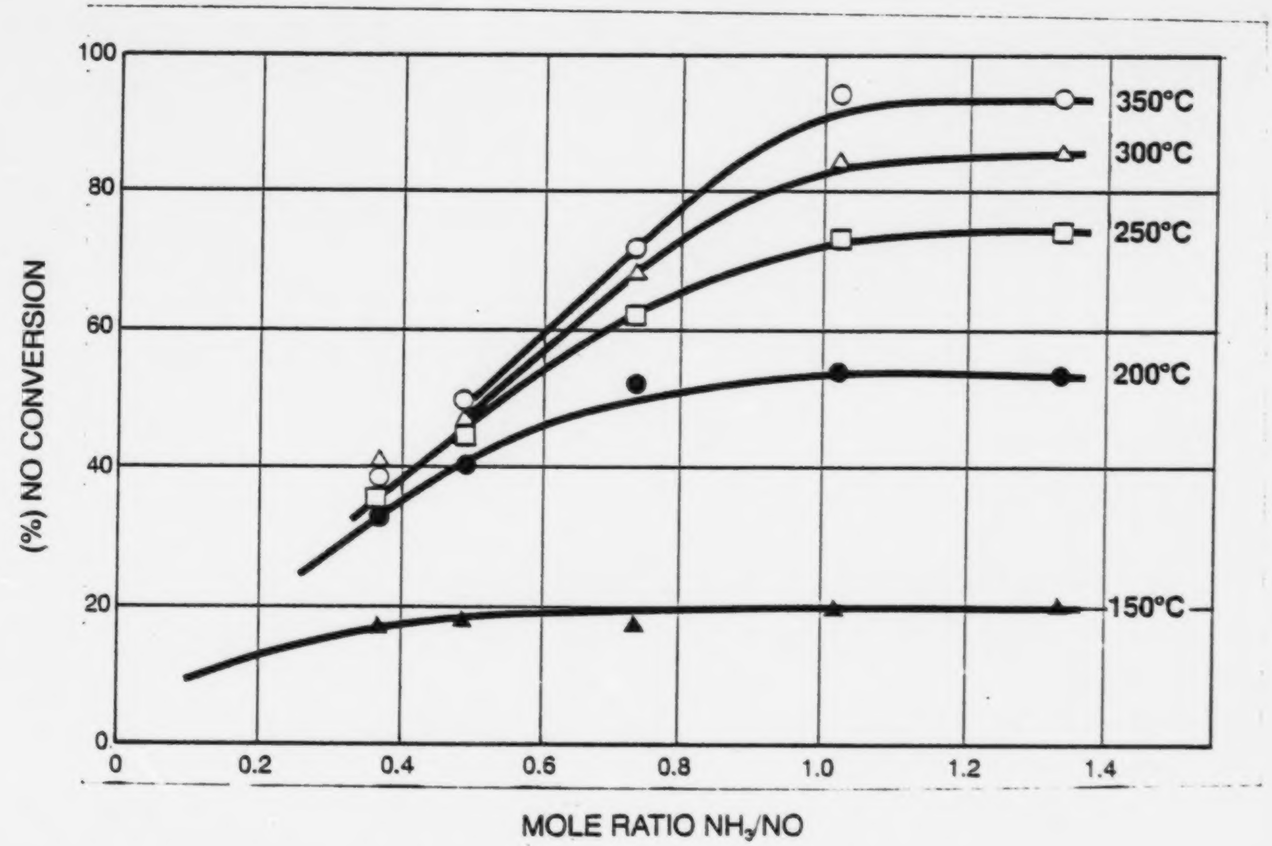
6% O_2

10% H_2O

10% CO_2

FIGURE 10

MOLE RATIO vs. NO CONVERSION



Catalyst: NOXNON 600

Area Velocity: 20.4 M^3/M^2-Hr.

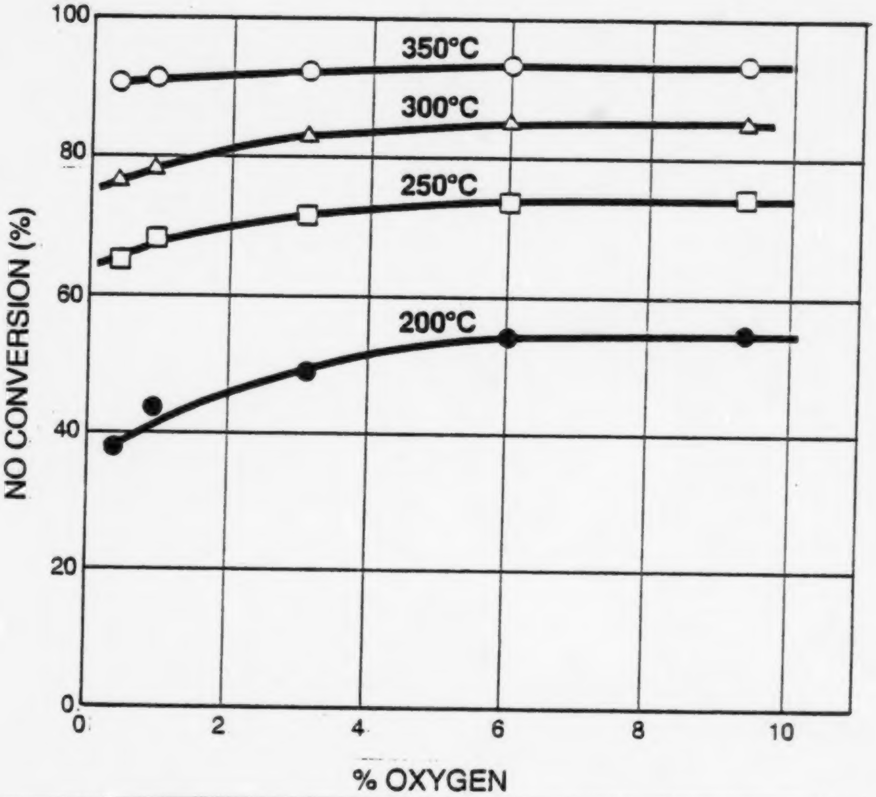
Gas Composition: 500 PPM NO

250 PPM SO_2

6% O_2, 10% H_2O, 10% CO_2

FIGURE 11

OXYGEN LEVEL vs. NO CONVERSION



Catalyst: NOXNON 600

Area Velocity: 20.4 M^3/M^2-Hr.

Gas Composition: 500 PPM NO
500 PPM NH_3
250 PPM SO_2
10% H_2O, 10% CO_2

TABLE 1

HITACHI ZOSEN
LIST OF COMMERCIAL PLANTS

	Customer	Treating Capacity, Nm^3/hr.	Flue Gas Source	Process	Completion
1	Osaka Gas Co., Sakai	53,000	LNG or naphtha-fired furnace	Ammonia Reduction	1975
2	Daiki Engineering, Chiba	5,000	LPG-fired furnace	Ammonia Reduction	1975
3	Idemitsu Kosan, Chiba	350,000	CO boiler and gas-fired heater	Ammonia Reduction	1975
4	Shin-Daikyowa Petrochemical, Yokkaichi	440,000	Fuel oil-fired boiler with wet-type desulfurization	Ammonia Reduction	1975
5	Hitachi Zosen, Osaka	6,000	Gas-fired annealing furnace	Ammonia Reduction	1975
6	Toshin Steel Mill, Himeji	70,900	Kerosene-fired steel heating furnace	Ammonia Reduction	1976
7	Kawasaki Steel, Chiba	762,000	Iron ore sintering plant with wet-type desulfurization	Ammonia Reduction	1976
8	Nippon Satetsu, Himeji	10,000	Fuel oil-fired steel heating furnace	Ammonia Reduction	1977
9	Kansai Oil Co. Sakai	150,000	Fuel oil fired boiler	Ammonia Reduction	1979

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BABCOCK-HITACHI NO_x REMOVAL PROCESS FOR FLUE GASES FROM COAL-FIRED BOILERS

By:

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Dr. F. Nakajima
Hitachi Research Laboratory of Hitachi Ltd.

ABSTRACT

In the previous symposium in 1978 we presented a paper titled "SOME EXPERIENCES OF NOx REMOVAL IN PILOT PLANTS AND UTILITY BOILERS". In that paper we stated the history of developments of our process, catalyst characteristics and several operating experiences. This time we intend to introduce some of the improvements and developments which we have achieved since then.

As far as coal-fired applications are concerned, there are two systems required. The first one is DeNOx with low dust loading where the DeNOx reactor is located downstream of the hot electrostatic precipitator (EP), and the second is DeNOx with high dust loading where the DeNOx reactor is located upstream of the cold EP. Although the selection of EP system should be determined mainly from the standpoint of performance of collecting fly ash through the boiler, our DeNOx process is applicable in either case.

As for the two commercial DeNOx plants with low dust loading from coal-fired boilers, we have already completed the design and manufacture, and they will go into commercial operation in November of 1980 and in July of 1981, respectively. Concerning the DeNOx with high dust loading, we will introduce in this paper the results of abrasion and performance tests under dust concentration of 15 to 20 g/Nm^3, through which we have confirmed the reliability of the catalyst.

Furthermore, another important thing in the Selective Catalytic Reduction (SCR) process is to reduce the conversion of SO_2 to SO_3 in order to minimize the influence on the downstream equipment. We have developed a catalyst with the lowest conversion rate less than 0.5 % without decreasing the NOx conversion activity at the rated load.

INTRODUCTION

Recently the use of coal as a substitute fuel for fossil oil has become of world wide importance. Since the characteristics of coal vary according to the location of mining, those of the coal-fired flue gas also vary. Generally, it contains not only more dust (15~25 g/Nm^3) than heavy oil-fired flue gas (0.05~0.2 g/Nm^3), but also higher NOx (Nitrogen Oxides) and higher SOx (Sulfur Oxides) concentration. This means that the DeNOx system, which is one of the environmental protection systems for coal-fired flue gas, demands more sophisticated technology than for heavy oil-, or gas-fired flue gas.

It was introduced at the previous symposium that Babcock-Hitachi had produced a high activity catalyst through extensive R & D, and has succeeded in developing it into a thin plate catalyst (a parallel flow type) for dirty gas. The plate type catalyst has been adopted for 20 plants out of 23 plants which have been ordered or are intended to be ordered soon. 10 of these plants are already operating satisfactorily or are under preoperation stage. Recently, demand for a DeNOx system for coal-fired flue gas has been increasing rapidly. Our first new DeNOx plant for low dust coal-fired flue gas (Hokkaido Electric Power Co., Tomato Atsuma P.S. Unit 1), the first in Japan to be installed in a new coal-fired P.S., will be in commercial operation this coming November, while the second plant (Electric Power Development Co. (EPDC) Takehara P.S. Unit 1) will also be in commercial operation next July. In addition to these, several plants including those for high dust flue gas are now being designed.

This paper describes the features of the DeNOx system for coal-fired flue gas in comparison with that for the oil-fired flue gas and includes an example of a DeNOx system for a coal-fired 500 MW boiler.

Furthermore, a pilot plant test using the plate type catalyst for a coal-fired flue gas has been conducted at Takehara P.S. on a co-study basis with EPDC.

ITEMS CONSIDERED IN THE DESIGN OF THE DeNOx SYSTEM FOR COAL-FIRED FLUE GAS

As the characteristics of coal-fired flue gas differ much from those of the heavy oil-fired flue gas, the peculiarities of the coal-fired flue gas have to be taken into consideration when designing the DeNOx system. Fig. 1 is a summary of the items considered and the measures taken in each item.

Our company has been producing the catalyst, which is a key point in the DeNOx technology, in accordance with our policy of manufacturing in our own works, from mixing of material to final product, under strict quality control since the start of development. Our success in developing a catalyst with high performance in practical use, is the result of a long program of catalyst improvement based on severe tests conducted in our research laboratory, factory, and in the field.

The catalyst, is composed of many thin plate elements, and its shape resembles the N.F elements of an air heater, so it is free from dust accumulation and has a low pressure drop. Additionally it has high erosion resistance and rigidity because it contains inner layer metal plate. It was confirmed that the performance of the catalyst had not decreased against the influence of SOx, halogen compounds (HCl, HF), and alkaline metals (K, Na) in the dust, even after 10,000 hours test with an actual coal-fired flue gas. Especially, the conversion ratio of SO_2 to SO_3 which is apt to cause problem in the downstream equipment is small enough to say that our DeNOx catalyst for coal-fired flue gas has a great advantage. Moreover, based on the results of model testing, improvements have been made to the reactor structure to prevent dust accumulation and non-uniform gas flow, and to simplify the loading and removal of catalyst.

SYSTEM FOR DeNOx

The flue gas treatment system for coal-fired boiler consists of three major systems, namely DeNOx, DeSOx and Dust Collection. Each system, in addition to fulfilling its own particular function, should, as part of the general boiler system, contribute to the total reliability and stability of the system over a long period of time, and be economical. Taking the above into consideration, we put two treatment systems for coal-fired flue gas into practical use. Those are shown in Fig. 2.

One is called a low dust DeNOx system, in which the boiler flue gas is first treated through a hot EP, and then the low dust flue gas is treated in the DeNOx reactor. The other is called a high dust DeNOx system, in which the boiler flue gas is treated in the DeNOx reactor without any pre-treatment after the economizer and followed by a cold EP.

The selection of the DeNOx system depends on the EP type which should be determined mainly from the standpoint of performance of collecting fly ash of which properties vary widely according to the location where fuel coal is mined. As far as DeNOx system is concerned, tests using actual flue gas have been conducted sufficiently for both systems, and it has been confirmed that both are practical as DeNOx system. The construction or the design of actual plants for each system is being carried out at present.

DeNOx SYSTEM WITH LOW DUST LOADING

We introduced some results of a low dust loading test for practical use at the previous symposium. We had performed a pilot plant test, in which we carried out long duration operation of more than 10,000 hours and confirmed various characteristics of the low dust loading. For instance, no change is found in DeNOx efficiency and reactor pressure drop, see Fig. 3, which shows that the DeNOx system can be operated quite stably. As shown in Fig. 3, DeNOx inlet dust concentration of 20, 100, 200 mg/Nm^3 were used and in addition 12 g/Nm^3 was experienced at hot EP trip, but there was no change in DeNOx efficiency nor reactor pressure drop. During this test the annual maintainance inspection, lasting 74 days, for the boiler unit was carried out and during this shutdown the DeNOx plant was exposed to the atmosphere, however this had no influence on the catalyst.

Summarizing the above, we can come to the conclusion that our DeNOx system has high reliability for practical use at low dust loading. The result of actual operation of the DeNOx system at Hokkaido Electric Power Co., and at plant of Electric Power Development Co. are expected to be excellent.

DeNOx WITH HIGH DUST LOADING

A high dust DeNOx system treats the flue gas from a boiler without any pretreatment. Therefore, the main aim in developing this system is to obtain an erosion resistant catalyst, which can withstand large amounts of, coarse, and hard dust composed of mainly silica and aluminum. For the purpose of overcoming this problem, an accelerated erosion test for the plate type catalyst was conducted with actual fly ash in Hitachi research laboratory. Fig. 4 shows the test equipment and Fig. 5 shows the evaluation of the test results.

According to our investigation of abrasion phenomenum, the erosion caused by abrasion with dust on the plate catalyst starts almost at the front edge of the plate, and then gradually proceeds downward. The catalyst material is integrally coated to stainless steel support plate which works as a protector against the abrasion by particulates. It was confirmed that our catalyst has extraordinary high erosion resistance. Summarizing the test results, the weight loss of the catalyst caused by abrasion can be estimated using the equation in Fig. 5.

Moreover, in order to conduct an abrasion test on the catalyst in an actual gas, the abrasion test catalyst was installed upstream of a hot E.P. in the same pilot plant as used for the low dust test mentioned in preceeding section. The test with high dust loading was conducted at a velocities of 4, 6, 8 m/s for about 3,000 hours. The result showed little or no erosion nor deactivity of the catalyst employed in the test. The result of this accelerated test with a gas velocity of 6 to 8 m/s was converted to the actual operating basis where the gas flows at a velocity of 4 to 5 m/s, and the result of this evaluation was that the operating duration would be equivalent to as much as 17,000 to 23,000 hours. Accordingly it is apparent that the catalyst can be used for commercial plant.

Fig. 6 shows the result of a high dust performance test, using the catalyst that had been used for the abrasion test mentioned above.

Up to now approximately 4,000 hours of performance testing, and as far as abrasion is concerned, a total of 7,000 hours of testing has been completed satisfactorily. It can be seen from Fig. 6 that little or no deactivity of the catalyst nor increase in pressure drop across the reactor has occured over the test duration, as was expected.

Several inquiries have been received regarding commercial DeNOx systems for high dust loading and some of them are at present under design.

LOW CONVERSION OF SO_2 TO SO_3

As illustrated in Fig. 1 when designing a DeNOx system for coal-fired flue gas, such points as "minimum effect on the downstream equipment" should be taken into consideration. To achieve this, the following conditions are essential.

1) A low conversion rate of SO_2 to SO_3
2) Minimized slip NH_3 from the DeNOx system

SO_3, mainly converted from SO_2, and NH_3 in the flue gas react to form NH_4HSO_4, as in the following reaction formula (1).

$$NH_3(g) + SO_3(g) + H_2O(g) \rightleftharpoons NH_4HSO_4(liq.) \qquad (1)$$

Fig. 7 shows the relation between the precipitation temperature of NH_4HSO_4 and SO_3 concentration with the parameter of NH_3 concentration. From Fig. 7, under the ordinary SO_3 and slip NH_3 concentration at reactor outlet, it is understandable that NH_4HSO_4 is formed at around 230 ~ 250 °C, which usually coincides with the temperature range in the intermediate section of the air heater downstream.

NH_4HSO_4 becomes such a sticky liquid over 150 °C that it is likely to adhere to the elements of the air heater and cause some dust to accumulate on the elements, ultimately resulting in problems, such as increase in pressure drop and plugging. In order to prevent this, it is preferable to slightly modify the air heater with regard to element shape, section deviding, and soot blowing procedure.

Most important is to minimize the formation of NH_4HSO_4, which causes trouble downstream. In order to prevent SO_3 and NH_3 from forming NH_4HSO_4, the SO_3 and NH_3 concentrations in the flue gas have to be kept as low as possible. To minimize slip NH_3, the reactor should be designed under an appropriate Space Velocity (SV), which is the gas quantity (Nm^3/h) divided by the catalyst volume (m^3), and operated with low $(NH_3)/(NOx)$ mole ratio, then the reaction between NH_3 and NOx should be maximum. The slip NH_3 can be kept at less than 5 ppm.

Regarding SO_3 which forms NH_4HSO_4, in addition to the SO_3 originated in the combustion flue gas, SO_3 is converted from SO_2 under the presence of the DeNOx catalyst. In the case of coal-fired gas, SO_2 is contained at the inlet of DeNOx reactor in such quantity, about 1000 ~ 2000 ppm, that large quantitis of SO_3 will be formed if we use an ordinary catalyst. It means 20 ~ 40 ppm of SO_3 will be formed newly with a catalyst which has a conversion ratio of SO_2 to SO_3 of 2 %, for instance. Needless to say the lowest conversion of SO_2 to SO_3 should be required for DeNOx catalyst for coal-fired flue gas.

Generally speaking, the catalyst which has high activity for NOx decomposition presented by formula (2) has also high conversion activity of SO_2 to SO_3 expressed in formula (3).

$$4NO + 4NH_3 + O_2 \rightleftharpoons 4N_2 + 6H_2O \qquad (2)$$

$$SO_2 + 1/2\ O_2 \rightleftharpoons SO_3 \qquad (3)$$

In other words if SO_2 oxidation activity is restrained, the NOx decomposing activity will also be greatly reduced. However, we succeeded in achieving low SO_2 oxidation activity without decreasing the NOx decomposing activity at boiler rated load by the selection of the proper catalyst component.

Fig. 8 shows the pilot plant test results using actual flue gas. During about 6,000 hrs operation conversion of SO_2 to SO_3 has been kept at less then 0.5 % without any tendency to increase.

DESIGN EXAMPLE FOR A 500 MW COAL-FIRED UTILITY BOILER

For reference, we would like to present an example of a design of a reactor for a 500 MW coal-fired utility boiler.
Design conditions are as follows.

Gas flow rate	1,500,000 Nm^3/h at MCR	
No. of reactors	2/boiler	
Gas temperature	370 °C	
NOx at inlet	500 ppm (5% O_2, dry basis)	
SOx at inlet	1,000 ppm	
Dust concentration	20 g/Nm^3	
DeNOx efficiency	Case-1	80 %
	Case-2	90 %
Slip NH_3	10 ppm (5% O_2, dry basis)	

The outline of the reactor designed, the shape of the catalyst and the catalyst loading procedure are illustrated in Fig. 9 and 10.

The basic dimensions of the catalyst element are about 1 mm thickness and 10 mm pitch. Gas from the economizer comes down, parallel to each catalyst plate, to avoid dust accumulation and two stages of catalyst bed are contained in the reactor. Generally soot blowers to clean the catalyst would be installed, however in the case of our plate type catalyst, which consists of thin catalyst plates arranged in parallel, very little dust accumulation on the catalyst was experienced, since there are very few corners in the flow path and the flow velocity is evenly distributed. In our design usually soot blowers for the catalyst are not necessary.

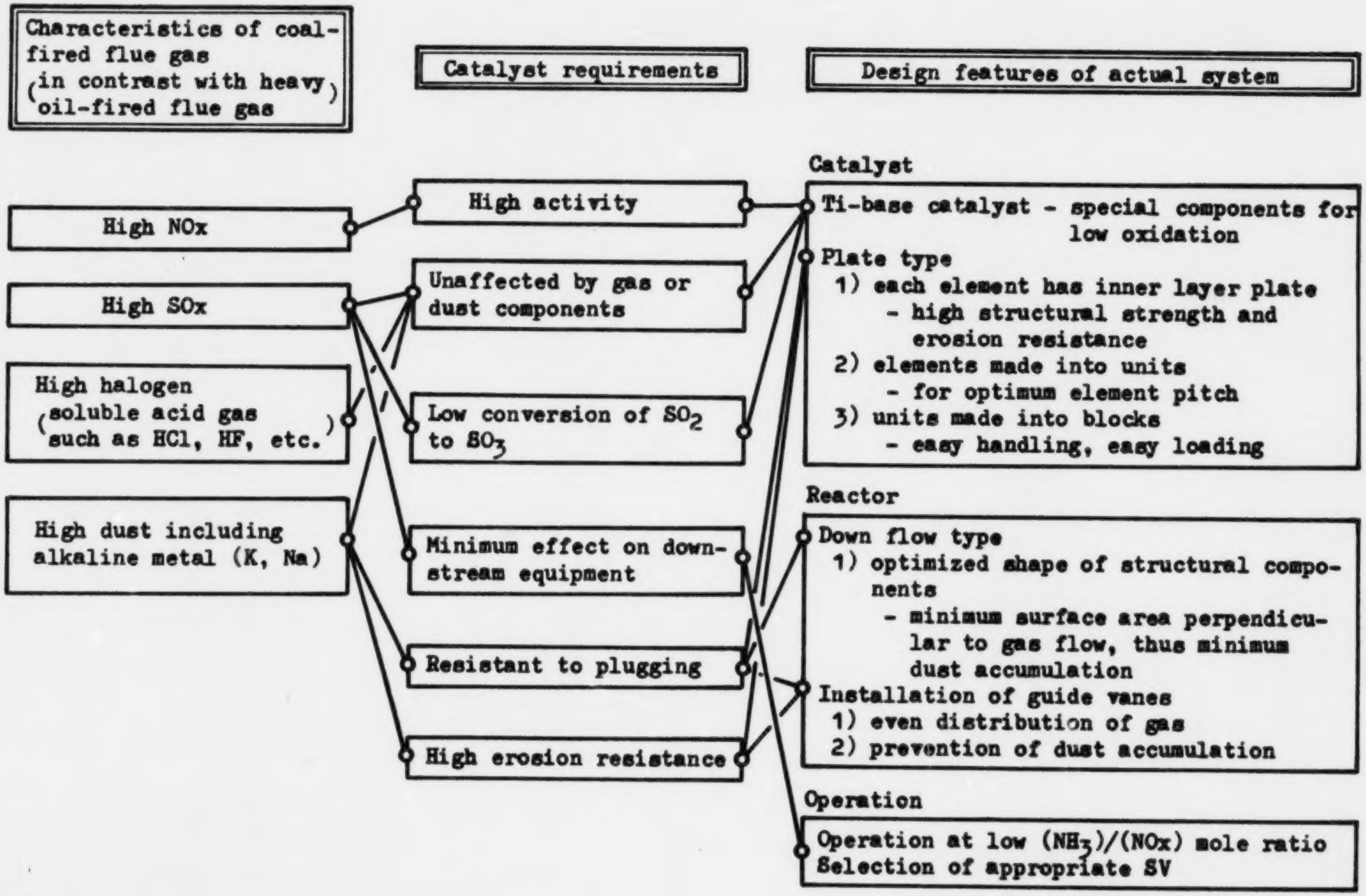
Fig. 11 shows the expected performance of the DeNOx for the boiler load at a DeNOx efficiency of 90 % (case-2). Generally, the DeNOx efficiency has a tendency to increase a little with lower load, which is caused by the relation of both the decreased gas flow rate and the decreased gas temperature. The pressure drop of a reactor can be designed to be as low as about 50 mmH_2O and the SO_2 oxidation ratio, to be as low as 0.3 ~ 0.5 %. Our study is based on a NOx concentration of 500 ppm at the DeNOx reactor inlet (boiler outlet), however, in Japan boiler combustion technology has improved so much that the NOx emitted by a new boiler can be less than 200 ppm.

SV is influenced by the reactor inlet NOx concentration, so that under the condition of constant DeNOx efficiency and slip NH_3, SV could be increased in proportion to lower NOx. The relation between SV and NOx concentration is shown in Fig. 12. Incidentally, if the NOx concentration at the DeNOx reactor inlet decreased from 500 ppm to 200 ppm, SV would increase by about 25 % and the quantity of catalyst required would decrease by about 20 %.

DeNOx efficiency is related to the quantity of catalyst, as shown in Fig. 13. It can be seen from Fig. 13 that the quantity of catalyst would decrease by about 30 %, in case the DeNOx efficiency lowered from 90 % to 80 %.

SUMMARY

In this paper we have related some of our recent studies with the Babcock-Hitachi dry catalytic NOx removal process. Regarding the DeNOx efficiency, we can obtain as high a value as we wish, by increasing the catalyst quantity, but the most important points for the DeNOx system in coal-fired plant are the catalyst characteristics of erosion resistance, anti-plugging and low conversion of SO_2 to SO_3. We believe that our plate type catalyst, which we manufacture in our own works under strict control, should satisfy all of those requirements. Hitachi Ltd., including Babcock-Hitachi, as not only a utility boiler manufacturer but also as an integrated machine manufacturer, highly experienced in the field, will continue to strive to improve this technology with all its technological resources.


Characteristics of coal-fired flue gas (in contrast with heavy oil-fired flue gas)
High NOx
High SOx
High halogen (soluble acid gas such as HCl, HF, etc.)
High dust including alkaline metal (K, Na)
Catalyst requirements
High activity
Unaffected by gas or dust components
Low conversion of SO2 to SO3
Minimum effect on down-stream equipment
Resistant to plugging
High erosion resistance
Design features of actual system
Catalyst
Reactor
Operation

Ti-base catalyst - special components for low oxidation

Plate type
  1) each element has inner layer plate
    - high structural strength and erosion resistance
  2) elements made into units
    - for optimum element pitch
  3) units made into blocks
    - easy handling, easy loading

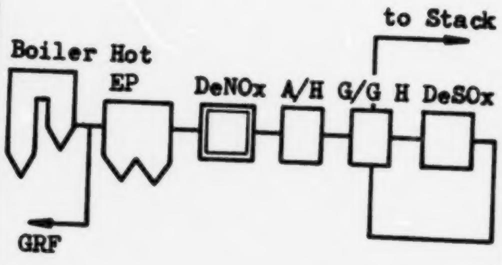
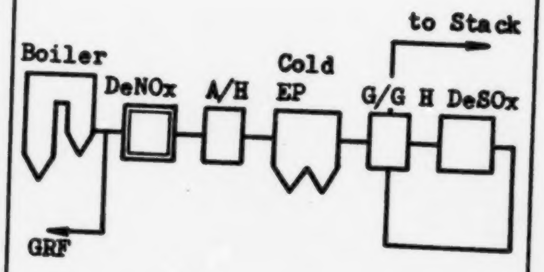
Down flow type
  1) optimized shape of structural components
    - minimum surface area perpendicular to gas flow, thus minimum dust accumulation

Installation of guide vanes
  1) even distribution of gas
  2) prevention of dust accumulation

Operation at low $(NH_3)/(NO_x)$ mole ratio
Selection of appropriate SV

Remarks : NOx (Nitrogen Oxides), SOx (Sulfur Oxides), SV (Space Velocity)

Fig. 1 Consideration on DeNOx System for Coal-Fired Flue Gas

Item	DeNOx with Low Dust Loading	DeNOx with High Dust Loading
System	 to Stack Boiler Hot EP DeNOx A/H G/G H DeSOx GRF	 to Stack Boiler DeNOx A/H Cold EP G/G H DeSOx GRF

Remarks : GRF (Gas Recirculation Fan) G/G H (Gas/Gas Heater)

Fig. 2 Flue Gas Treatment System for a Coal-Fired Boiler

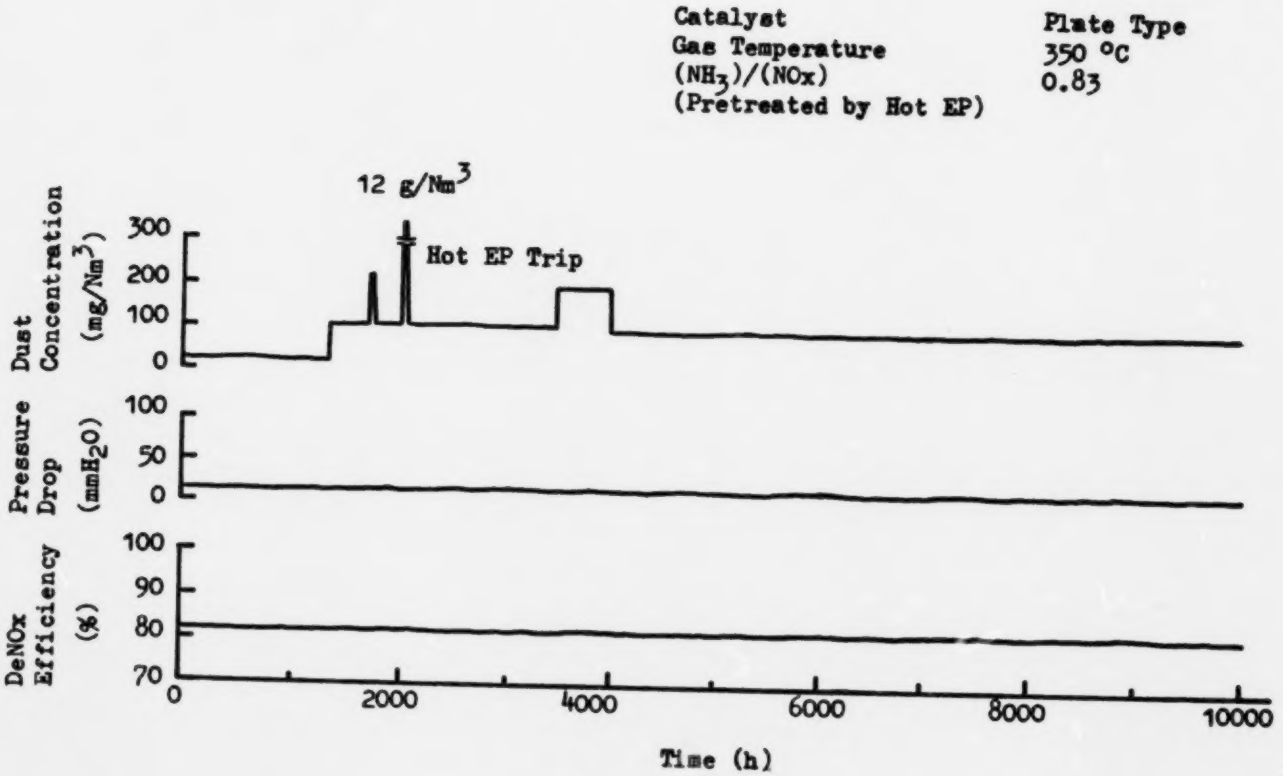


Fig. 3 Pilot Plant Test of Parallel Flow Reactor Treating Flue Gas from Coal-Fired Utility Boiler

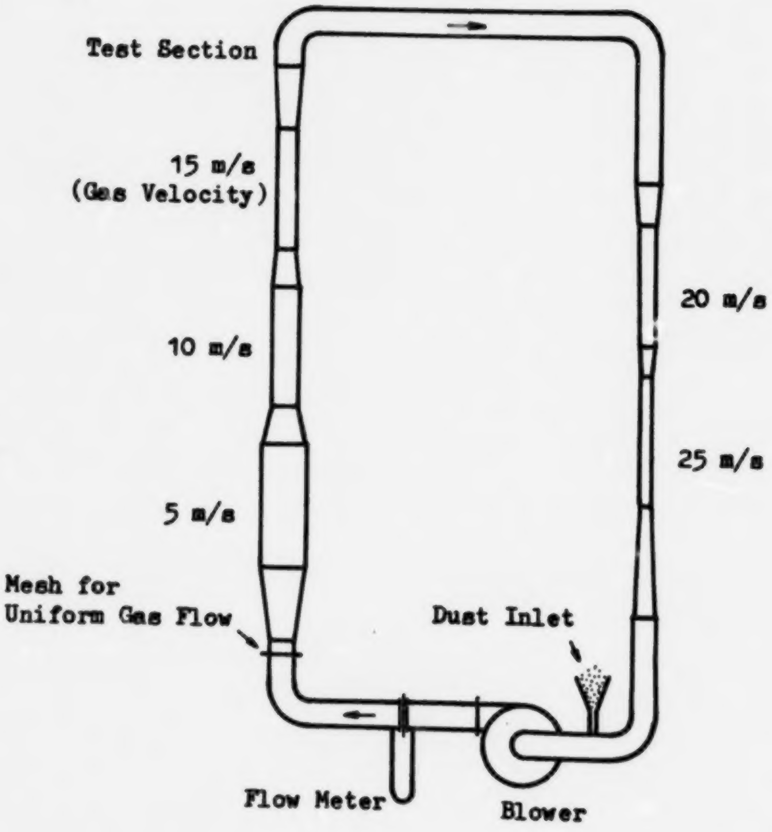

Test Section
15 m/s
(Gas Velocity)
10 m/s
5 m/s
Mesh for
Uniform Gas Flow
Flow Meter
Blower
Dust Inlet
20 m/s
25 m/s

Fig. 4 Equipment for Catalyst Abrasion Test

Experimental Equation

$$W = K \cdot V^{2.9} \cdot C^{1.7} \cdot T^{0.64}$$

W : Weight Loss of Catalyst (wt%)
K : Constant
V : Gas Velocity (m/s)
C : Dust Concentration (g/Nm^3)
T : Operating Time (h)

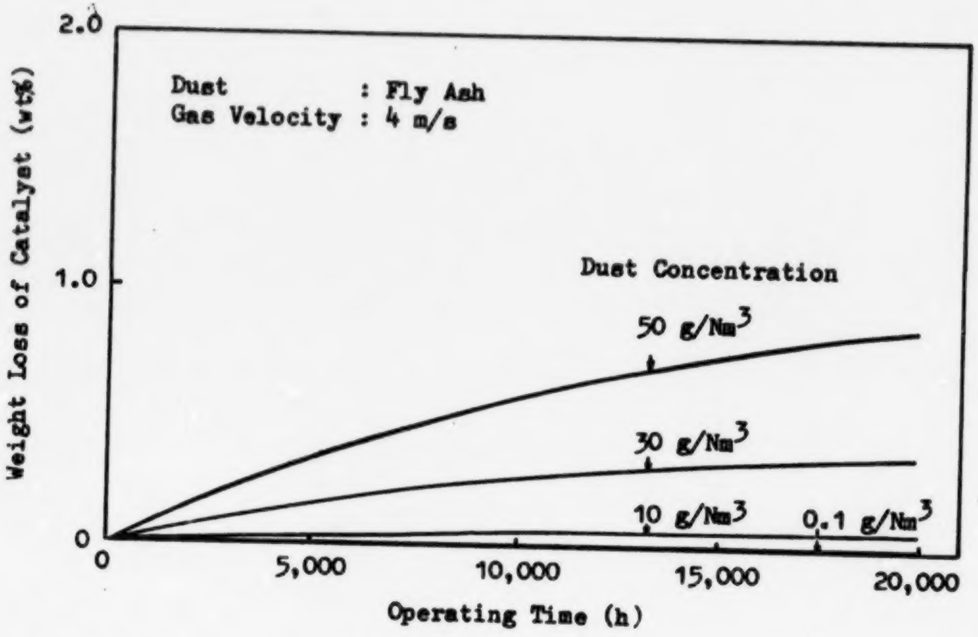


Fig. 5 Anticipated Weight Loss of Plate Type Catalyst vs Operating Time

Test Purpose	Abrasion Test	Performance Test
Gas Temperature	346 ~ 360 °C	348 ~ 353 °C
NOx	244 ~ 345 ppm	285 ~ 472 ppm
SOx	1100 ~ 1700 ppm	(1100 ~ 1700) ppm
Dust Concentration	16.1 ~ 19.0 g/Nm^3	17.3 ~ 20.2 g/Nm^3
LV	-	4 m/s
$(NH_3)/(NOx)$	0	0.83

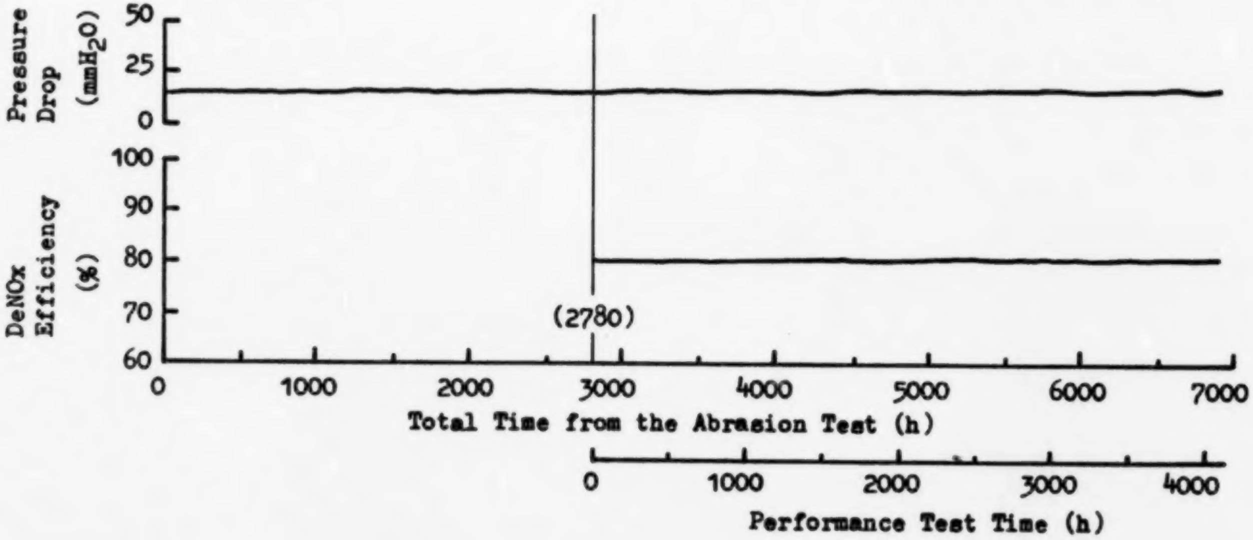


Fig. 6 Operation Chart of the Parallel Flow Type DeNOx Pilot Plant for Coal-Fired Boiler Flue Gas

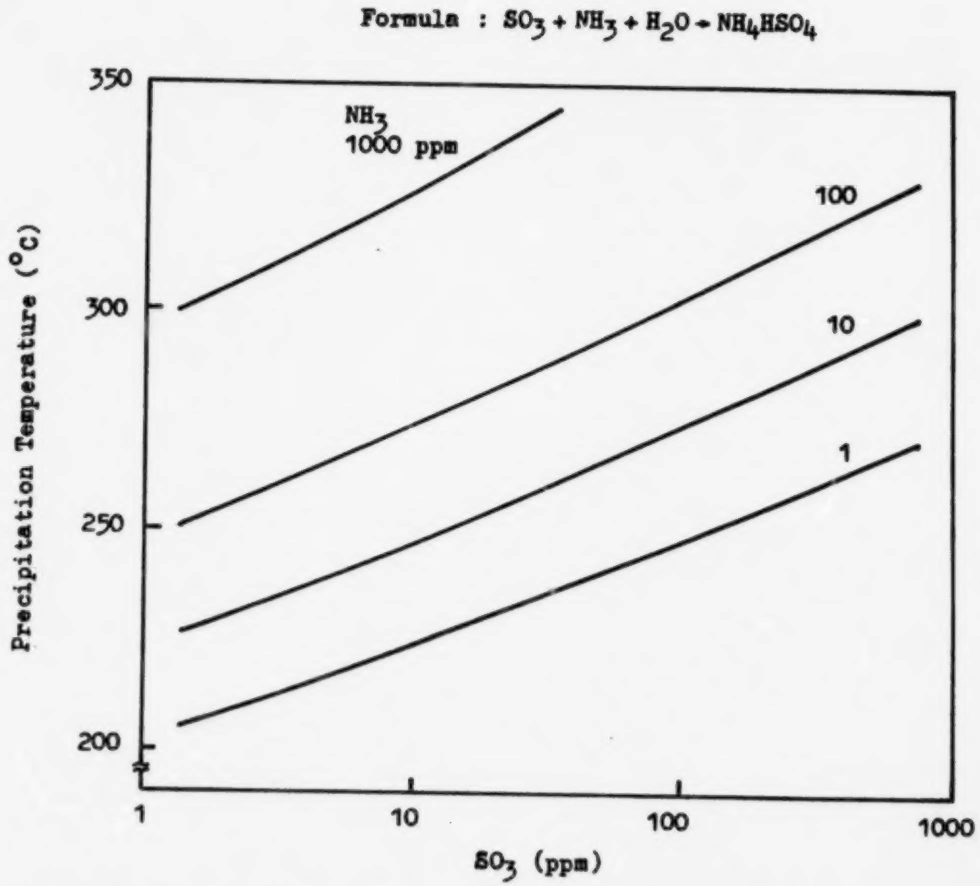


Fig. 7 Precipitation Temperature of Ammonium Bisulfate (NH_4HSO_4)

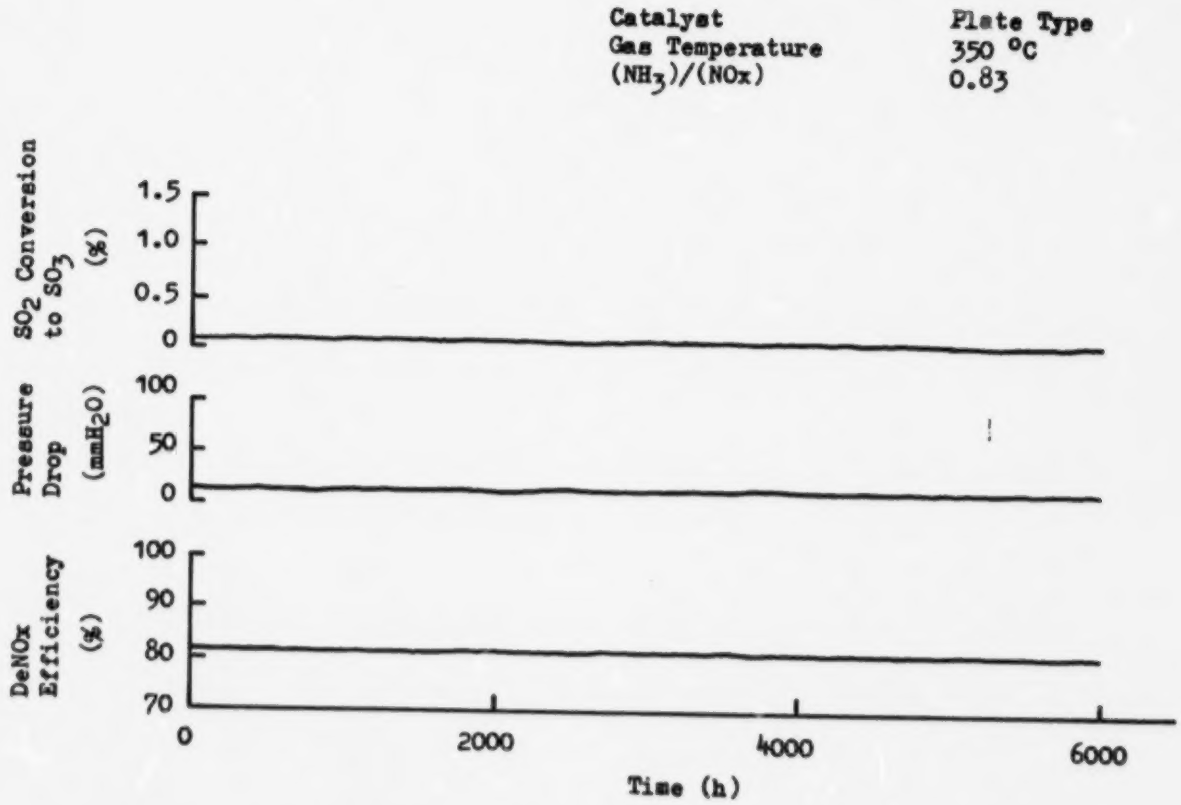


Fig. 8 Pilot Plant Test of a Parallel Flow Reactor Treating Flue Gas from a Coal-Fired Utility Boiler

(m)

	CASE-1	CASE-2
W	11	11
L	13	13
H	18	19

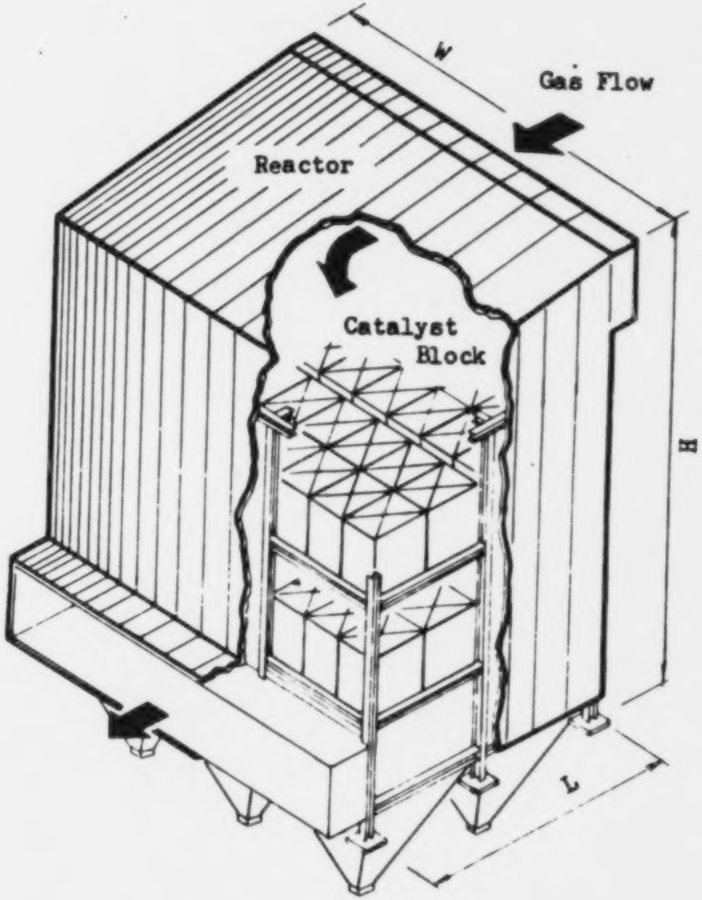

W
Gas Flow
Reactor
Catalyst
Block
H
L

Fig. 9 Outline of Reactor for 500 MW Coal-Fired Boiler

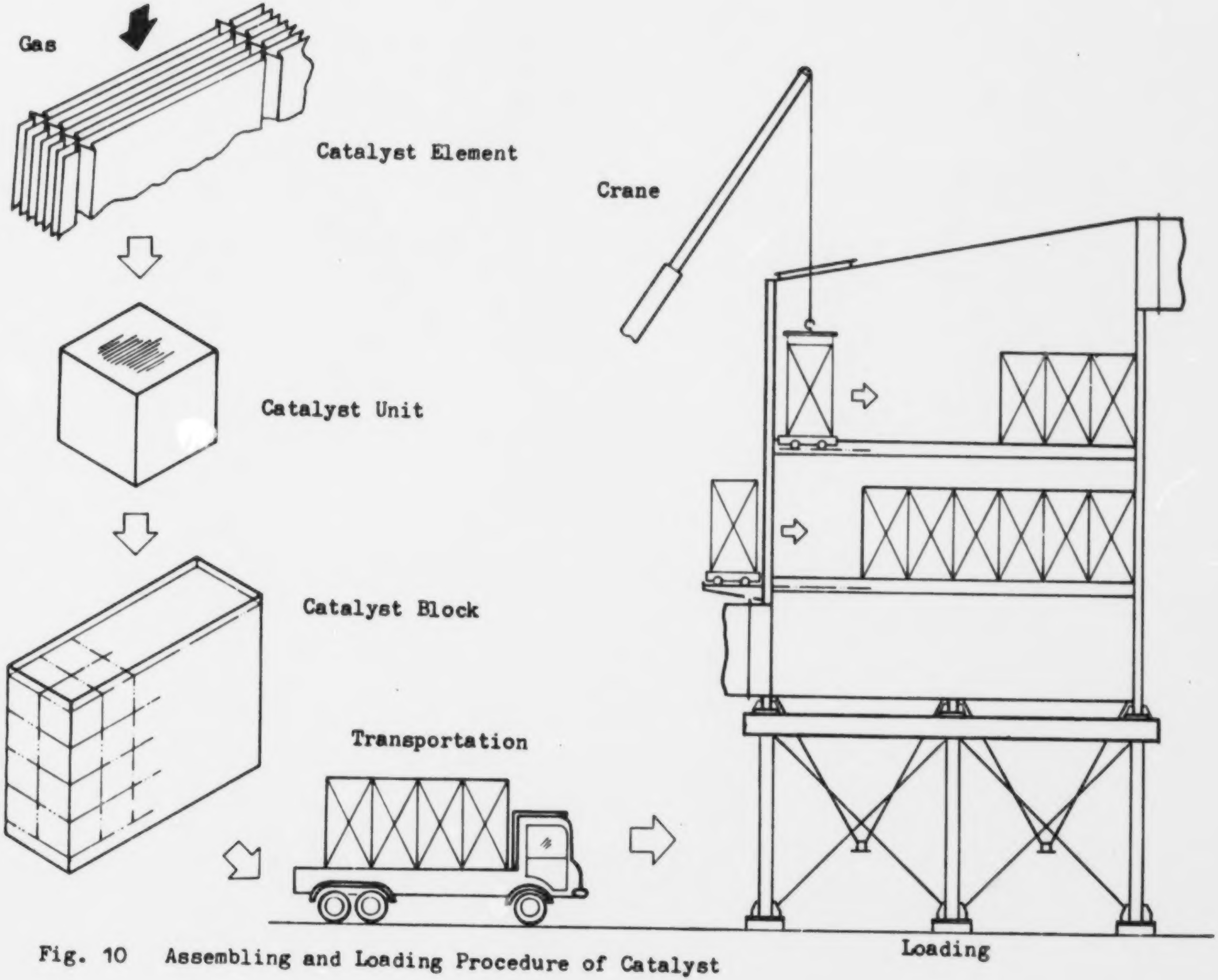

Gas
Catalyst Element
Crane
Catalyst Unit
Catalyst Block
Transportation
Loading

Fig. 10 Assembling and Loading Procedure of Catalyst

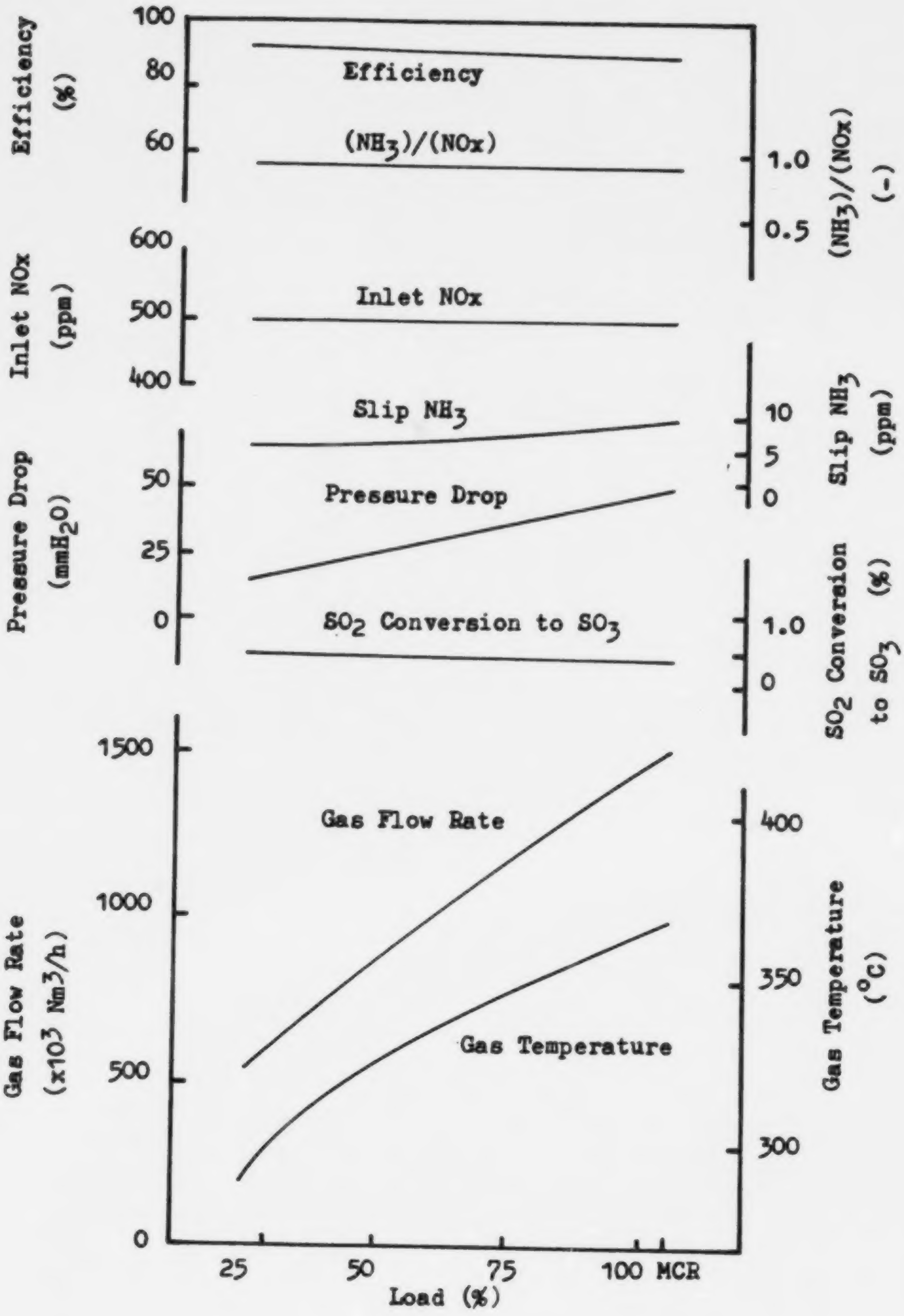


Fig. 11 Expected Performance with Load Conditions

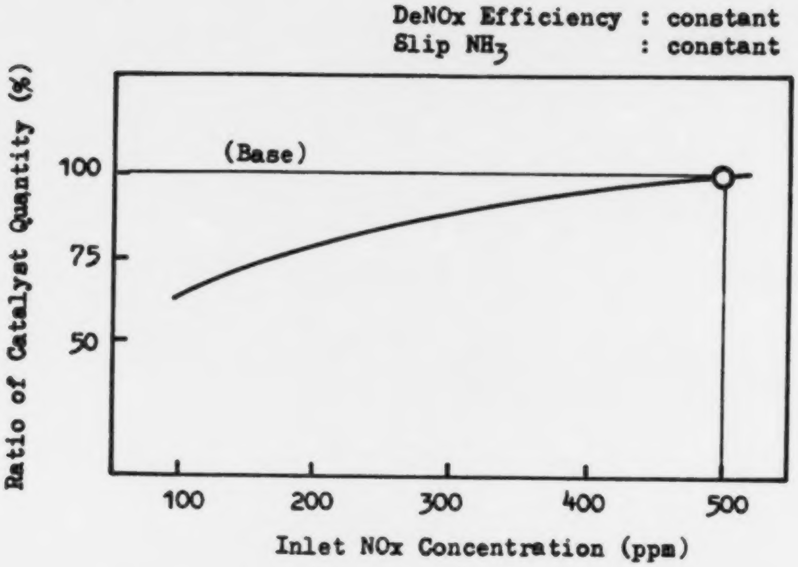


Fig. 12 NOx Concentration vs Catalyst Quantity

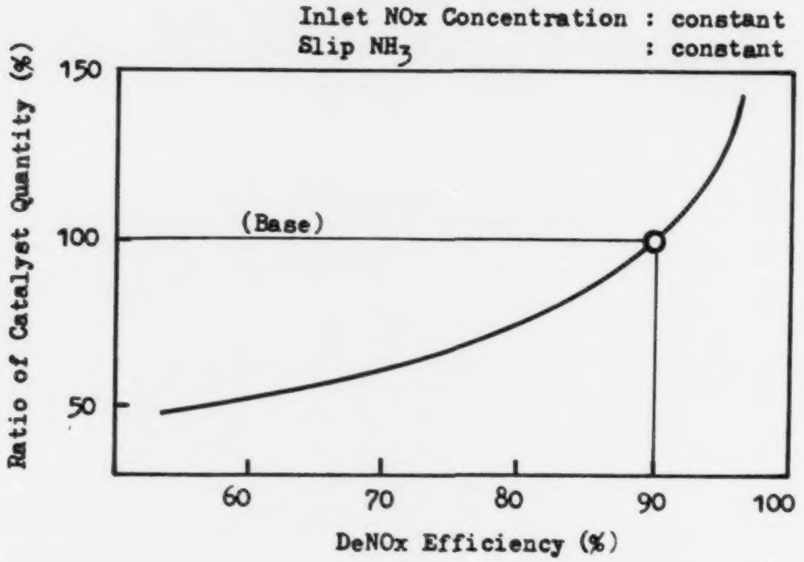


Fig. 13 DeNOx Efficiency vs Catalyst Quantity

TEST SUMMARY OF AN INTEGRATED FLUE GAS TREATMENT SYSTEM

Utilizing the Selective Catalytic Reduction Process for a Coal-Fired Boiler

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ABSTRACT

This research program was initiated to investigate whether denitrification of flue gases from a coal-fired boiler can be effectively accomplished by using the selective catalytic reduction process in combination with desulfurization and dust control as an integrated system.

An experimental system was designed to handle between 1000 and 2000 NM^3/hr of flue gas and measure such parameters as catalyst life, catalyst plugging, catalyst abrasion rate due to ash, air heater plugging, hot ESP, wet ESP, and bag filter efficiencies, along with the efficiency of a limestone desulfurization system.

The results of this testing have shown that this integrated approach to flue gas cleanup is feasible and may be incorporated into a full-scale, coal-fired boiler flue gas design. This testing will be continued to develop a more reliable integrated flue gas treatment system prior to commercialization.

SECTION 1

INTRODUCTION

Recent attempts to substitute coal for oil as an energy source for power plants have created a decrease in the overall air quality. To correct this trend, the need for an integrated flue gas treatment system has become apparent. This new integrated system must reduce the concentrations of dust, SO_x, and NO_x in the flue gases below those created by the former use of heavy oil.

To meet these needs, IHI started a joint research and development project for an integrated coal flue gas treatment system with Electric Power Development Co., Ltd. To prepare for this project, IHI started denitrification testing in January 1978 on flue gas from a coal-fired boiler at Isogo Power Station of Electric Power Development Co., Ltd. In December 1978 a desulfurizing system was installed on the downstream side of the denitrification system. Various tests were conducted concerning denitrification, desulfurization, and dust collecting functions of this integrated flue gas treatment system. These tests have provided the opportunity to analyze the problems and possible solutions concerning the eventual commercialization of this integrated flue gas treatment system.

Foster Wheeler is presently marketing the IHI Selective Catalytic Reduction Process in the United States.

SECTION 2

TEST OBJECTIVE

The individual techniques to treat air pollution emissions from coal-fired boilers, such as NO_x, SO_x, and dust, have been well established. However, these tests were aimed at integrating these techniques into a total system, including waste water treatment. The following discussion is concerned with various problems and their solutions regarding such an integrated flue gas treatment system.

The major test objectives were as follows:

1. Determining $DeNO_x$ catalyst life
2. Preventing the catalyst layer from plugging with ash
3. Preventing the abrasive wear on the catalyst due to ash
4. Detecting ways to prevent plugging of the gas air heater (GAH)
5. Developing of a low SO_3 conversion ratio catalyst
6. Determining dust collecting efficiencies of a high temperature electrostatic precipitator, bag filter, and a wet type electrostatic precipitator
7. Measuring characteristics of the desulfurization system, such as efficiency of removal, effect of dust, quality of gypsum, method of waste water treatment, and ways to prevent plugging of the reheat system.

SECTION 3

TEST SYSTEM

Two DeNO$_x$ systems were selected as the best available for this testing. The first was chosen to handle the high dust condition directly from the boiler and located ahead of the air heater and cold ESP. The other DeNO$_x$ system was installed between the hot ESP and the air heater to handle the low dust condition. (Reference Figure 1 on the following page.) In this test, the flue gas reheater was a standard design, and the FGD system used a common wet limestone process.

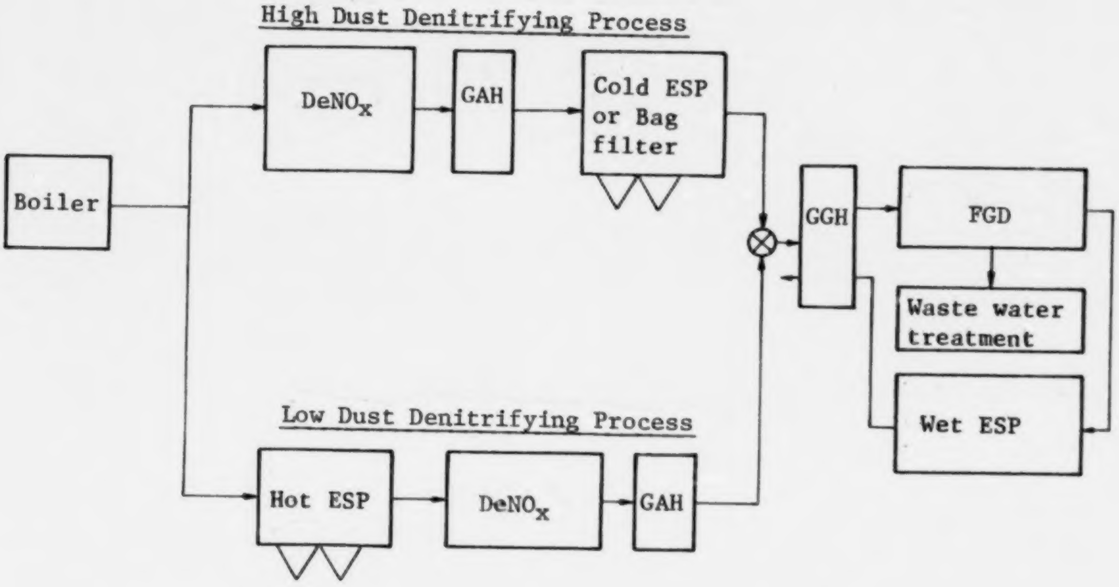

High Dust Denitrifying Process
DeNO$_x$
GAH
Cold ESP or Bag filter
Boiler
GGH
FGD
Waste water treatment
Wet ESP
Low Dust Denitrifying Process
Hot ESP
DeNO$_x$
GAH

FIGURE 1

224

SECTION 4

SPECIFICATIONS OF TEST EQUIPMENT

The specifications of the major test equipment used for this program are shown in Table I below, and Figure 2 on following page shows the Schematic Flow Diagram.

TABLE I

Test Equipment	Specifications		
Denitrification device	Denitrifying method	Selective catalytic reduction process using NH_3	
	Type of reactor	Vertical downward flow type fixed bed	
	Shape of catalyst	Square honeycomb type catalyst	
	Capacity	1000 NM^3/h x 2 for high dust 1000 NM^3/h x 2 for low dust	
		High Dust	Low Dust
Air heater (GAH)	Type	(Regenerative type) High temperature element: DU Medium and low temperature element: NF	(Generative type) NF-Single layer
	Capacity	2000 NM^3/h x 1	2,000 NM^3/h x 1

TABLE I (cont'd.)

Test Equipment	Specifications			
		High temperature electrostatic precipitator	Bag filter	Wet electrostatic precipitator
Dust collector	Type	Dry horizontal gas flow type	Reverse washing bottom inlet type	Wet type flat plate
	Capacity	2000 NM^3/h	2000 NM^3/h	1000 NM^3/h
Reheater (GGH)	Type	(Generative type)		
	Capacity	1000 NM^3/h x 1		
Desulfurization device	Type	Limestone wet process FGD system (Both separation and mixing of ash are possible)		
	Capacity	1000 NM^3/h x 1 Dust removing tower, absorption tower, mist eliminator, gypsum recovery process, waste water treatment equipment		

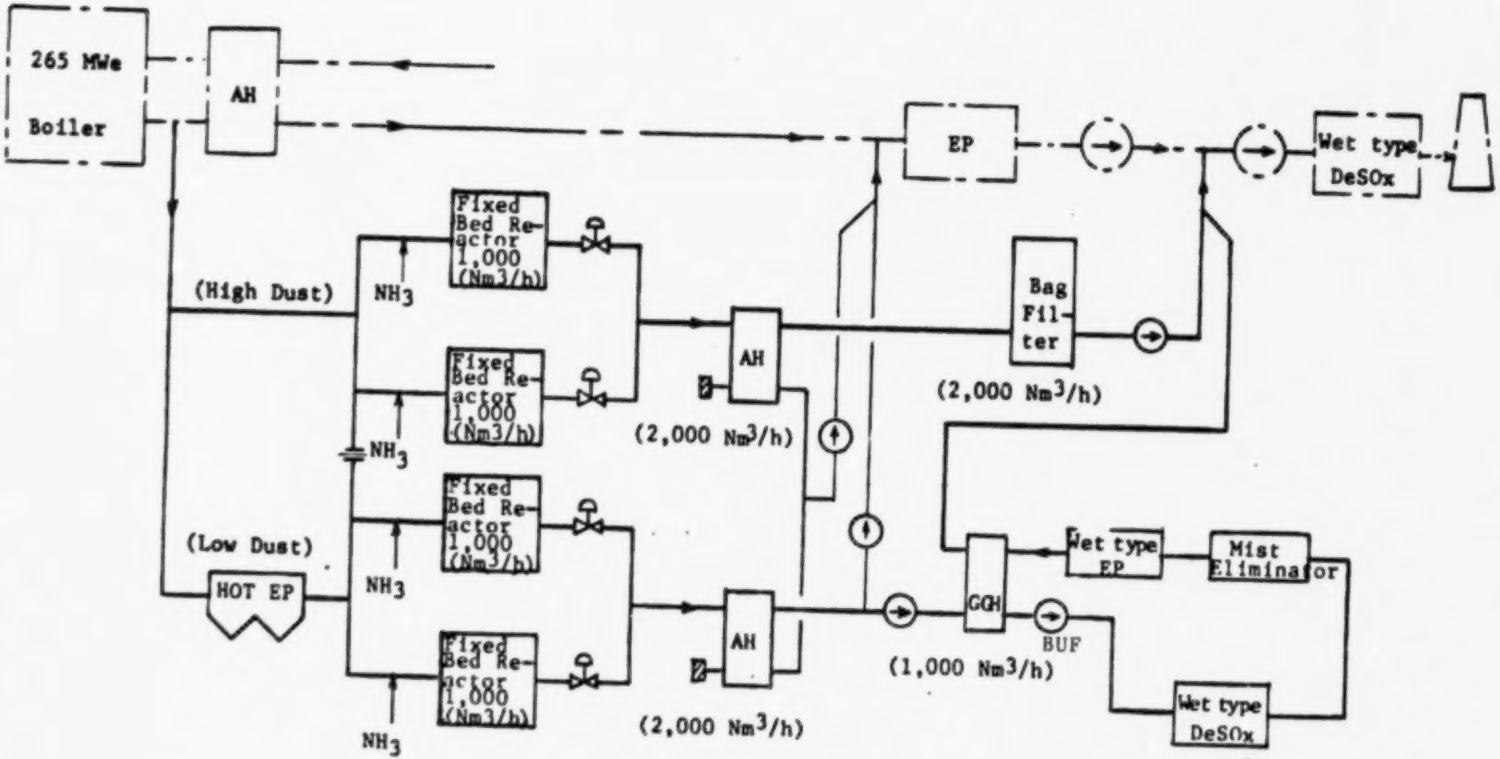

265 MWe
Boiler
AH
EP
Wet type DeSOx
(High Dust)
(Low Dust)
HOT EP
NH3
NH3
NH3
NH3
Fixed Bed Reactor 1,000 (Nm3/h)
Fixed Bed Reactor 1,000 (Nm3/h)
Fixed Bed Reactor 1,000 (Nm3/h)
Fixed Bed Reactor 1,000 (Nm3/h)
AH
AH
(2,000 Nm3/h)
(2,000 Nm3/h)
Bag Filter
(2,000 Nm3/h)
GGH
BUF
(1,000 Nm3/h)
Wet type EP
Mist Eliminator
Wet type DeSOx
(1,000 Nm3/h)

Fig. 2 Schematic Test flow diagram of integrated flue gas treatment systems

SECTION 5

TEST SCHEDULE

This test and research project was initiated as a result of early DeNO$_x$ testing that started January 1968. It soon became apparent that there was a need to combine NO$_x$, SO$_x$, and dust removal in one integrated system and that this combined system looked viable based on the early individual testing. Table II shows the schedule of the testing from 1977 through 1980.

TABLE II

	1977	1978	1979	1980
Installation work	Denitrification device High Temperature ESP, GAH	Desulfurization device	Bag filter	
$DeNO_x$ test		$DeNO_x$ test (High/Low dust) • Test to check the construction of the reactor • Performance test • Test to check pitch of honeycomb mesh • Development of low SO_3 conversion ratio catalyst • Measures to prevent plugging of GAH • Measures to prevent plugging and wear of catalyst		
Integrated flue gas treatment test			Integrated flue gas treatment test • Life test of catalyst (Low dust system, High dust system) • Desulfurizing test Measures to prevent plugging of GAH and GGH • Long-term test to check plugging of GAH and GGH • Life test of bag filter	

SECTION 6

DISCUSSION OF TEST RESULTS

The results of this integrated flue gas treatment test can be summarized as follows:

DENITRIFICATION EQUIPMENT

Life Test of Catalysts

Tests to determine the life of the catalyst are now in progress. Some catalysts have passed 9,000 hours during low dust conditions and 7,000 hours for high dust conditions. No significant deterioration was observed under either of these conditions, and the catalysts have shown that a stable $DeNO_x$ efficiency of greater than 80% is possible. These tests will be continued in order to test their performance over a longer period.

Figures 3 and 4 on the following pages show the life test results of catalyst at low and high dust conditions.

Plugging of Catalyst Layer

At the high dust conditions, no deposits or ash accumulation were observed on the catalyst. While at the low dust conditions, the ash has a smaller grain size and is more prone to sticking to the catalyst without the benefit of having the larger particles perform a self-cleaning action.

As a result of this non-self-cleaning action, partial deposits or accumulations of ash were observed in this case. It was shown, however, that the plugging of the catalyst layer can be prevented by considering such factors as the flue gas flowrate, mesh size of honeycomb catalysts, catalyst support construction, and adoption of a catalyst layer using seamless catalyst in the flow direction.

Place of test		Isogo Power Plant	
Catalyst		Square mesh honeycomb type catalyst (A-III type)	
Condition	Concentration of dust	g/Nm³	12 ~ 20
	Concentration of SOx		300 ~ 900
	Reaction temperature	°C	350
	NH_3/NOx	Mol ratio	0.9
	SV value	h^{-1}	3,300

(800 ~ 900ppm after Feb. 14, 1980)

Fig. 3 Results of test for the concentration of high dust

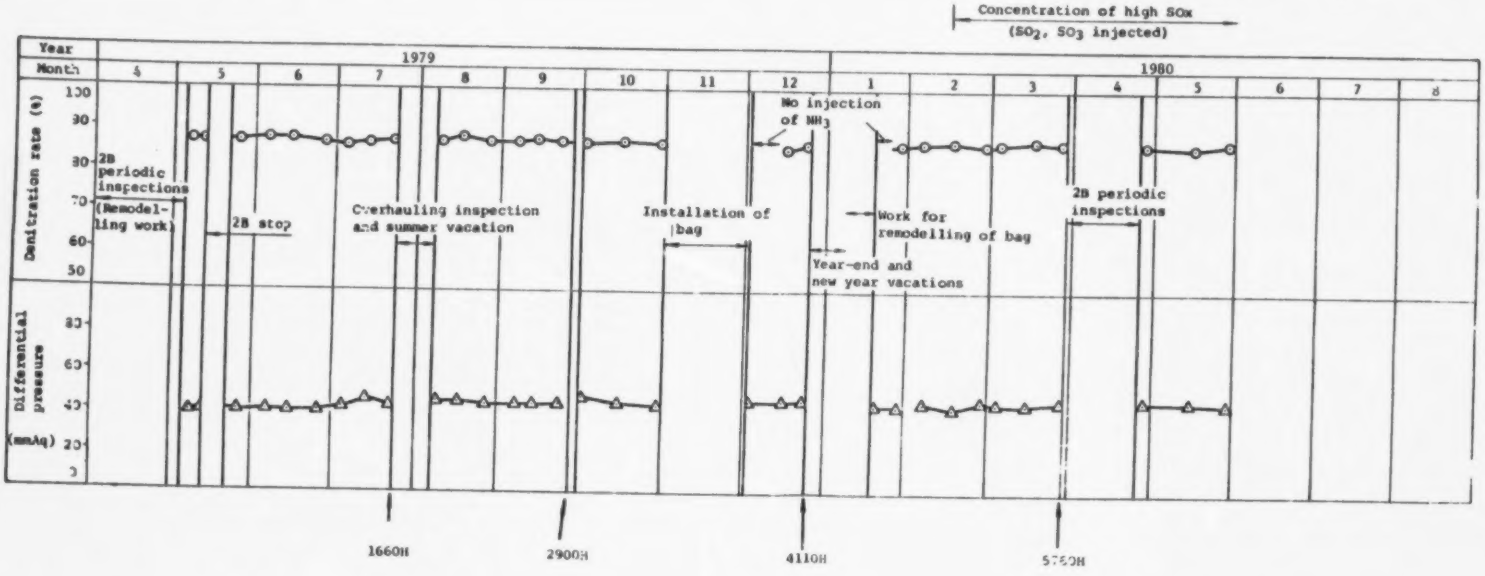
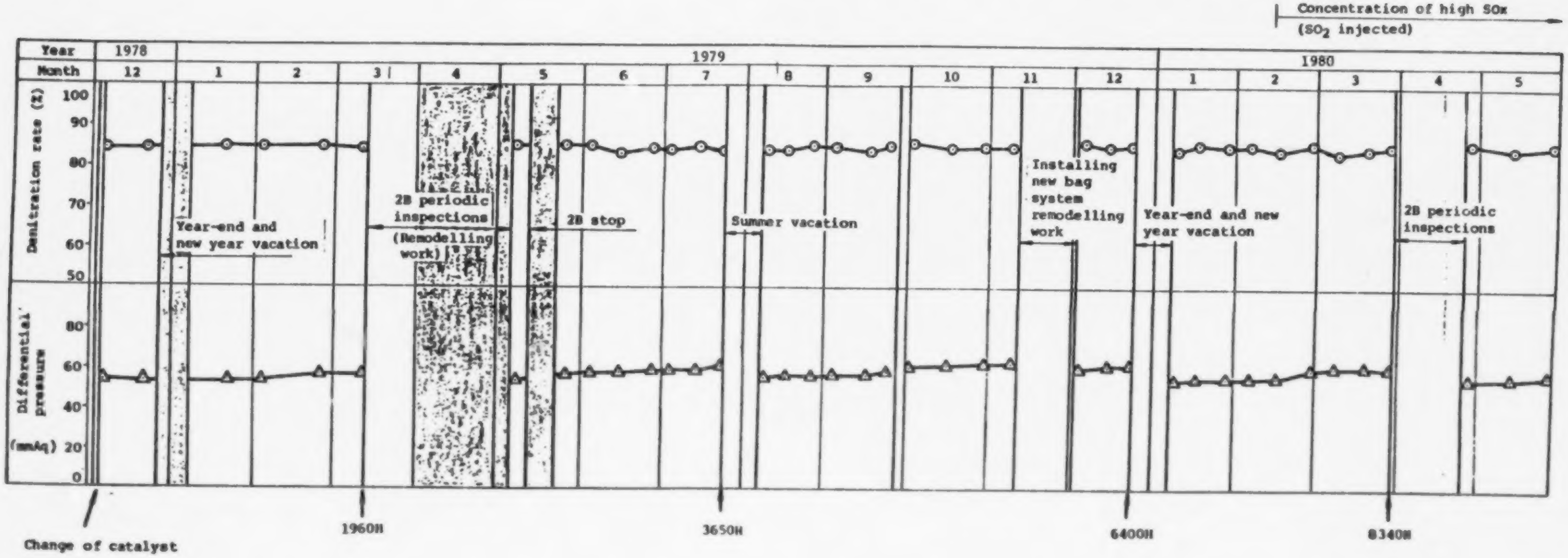


Fig. 4 Results of test for the concentration of low dust

Place of test		Isogo Power Plant	
Catalyst		Square mesh honeycomb catalyst (A-II type)	
Condition	Concentration of dust	g/Nm^3	0.1 ∿ 0.2
	Concentration of SOx		300 ∿ 900
	Reaction temperature	°C	350 °C
	NH$_3$/NOx	Mol ratio	0.9
	SV value	h^{-1}	2,800

(800 ∿ 900ppm after Feb. 14, 1980)



Erosion of Catalyst

The most significant factor which affects the life of the catalyst in coal-fired applications is the abrasive wear caused by the ash in the combustion gas. However, these tests have shown that this wear can be effectively controlled by designing for the following conditions: selecting an appropriate flowrate for gas, choosing a catalyst having a high resistance to wear, and eliminating the seams in the catalyst in the direction of gas flow. The results of the test for the 7,000 hr catalyst life test showed no significant wear during inspection when the above design considerations were taken.

During this series of testing, an accelerated wear test was conducted over a relatively short period of time, where the flowrate of gas and the concentration of dust were extremely high when compared with those at normal conditions.

The result of this test showed the abrasive wear of the catalyst to be calculated as 0.1mm/year, indicating that the catalyst can be used effectively for several years at normal conditions. At low dust conditions, the concentration is 1/100 of that under high dust conditions and the grain size is smaller, thereby eliminating the concern about abrasive wear of the catalyst.

Development of Low SO_3 Conversion Ratio Catalyst

The rate of conversion at which parts of SO_2 contained in the gas are converted into SO_3 in the catalyst layer affects the equipment on the downstream side through corrosion or plugging. However, it has been shown through research that a catalyst having an extremely low conversion rate of SO_2 to SO_3 of less than 1% can be manufactured.

PLUGGING OF GAS AIR HEATER (GAH) ELEMENTS

This plugging, caused by an NH_3/SO_3 based product consisting of unreacted NH_3 from the $DeNO_x$ process and SO_3 contained in the flue gas, is one of the problems that emanate from the operation of the $DeNO_x$ process. However, this problem can be solved by considering the design of the heater element, soot blowing design, and frequency of soot blowing.

Where there is a high dust concentration in the flue gas, a large cleaning effect from the dust can be expected so that the GAH can be normally operated by using the soot blower several times a day.

In the case of a low dust concentration flue gas, a cleaning effect from the dust cannot be expected, thus creating a greater possibility for the heater elements plugging. However, even for this low dust condition, plugging of the elements can be prevented by designing the element shape to produce an improved overall soot blowing effect. For example, according to the results of a test conducted for over 3,000 hours, close inspection showed no significant plugging of heater elements. This test is still in progress in order to check whether it is possible to operate continuously over a long period of time using these design improvements.

Additional test results showed no significant corrosion or decrease in the wall thickness of the heater element for either the low or high dust conditions.

Figure 5 shows the results of pressure loss testing for the GAH for both high and low dust conditions.

DUST COLLECTOR

High Temperature Electrostatic Precipitator (Hot ESP)

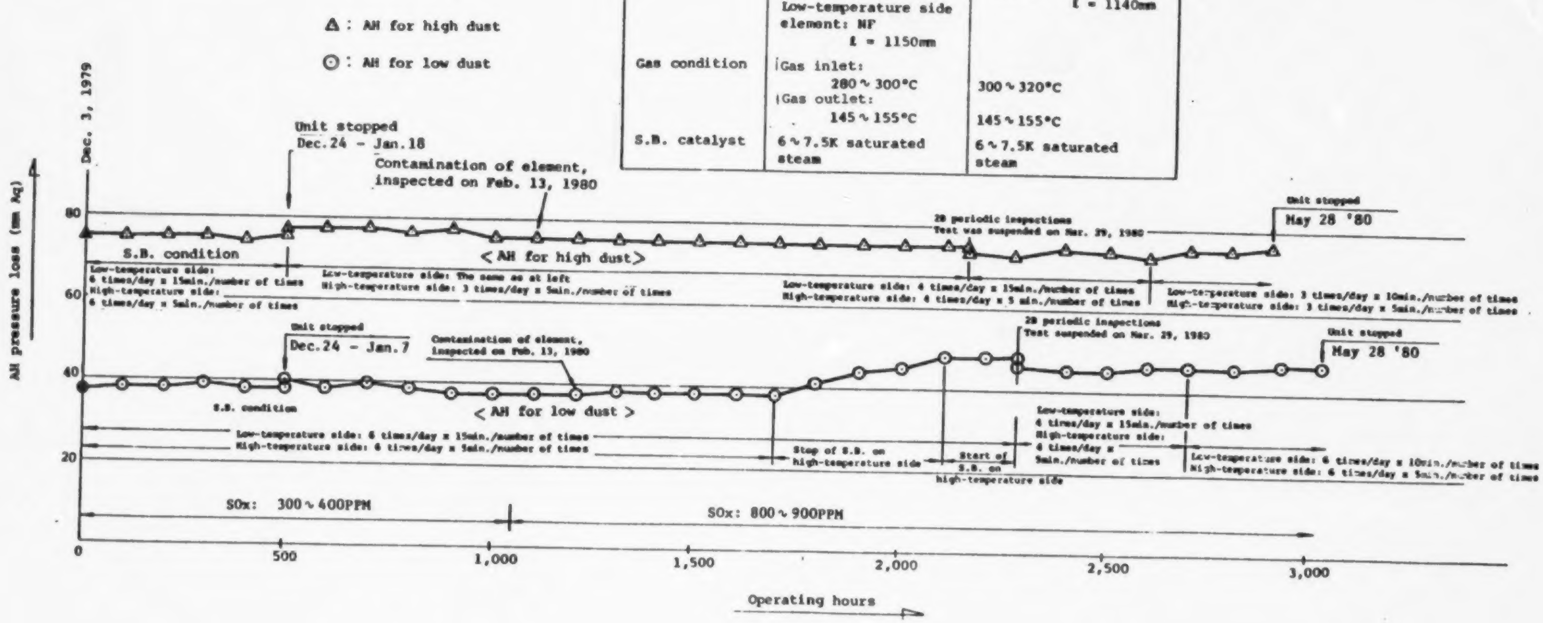
Currently there are various opinions as to the efficiency of the hot ESP. According to the results of these tests, the collection efficiency of a hot ESP proved highly effective on dust in the flue gas from coal containing 0.6% sulfur.

Wet Type Electrostatic Precipitator (Wet ESP)

At present, the concentration of dust at the outlet of a desulfurization system has to be about 20 mg/NM^3 in Japan. However, in order to meet more strict requirements in the future, a wet ESP was installed on the outlet side of the desulfurization device used for this testing. The next test series showed the dust collection efficiency of wet ESP to be about 93%. These results show that the concentration of dust at the outlet of the FGD unit can be maintained under 10 mg/NM^3 when this wet ESP is used, even if the scale effect is taken into consideration.

Fig. 5 |Pressure loss in GAH

Class. / Item	AH for high dust	AH for low dust
Consturction of AH	High-temperature element: DU ℓ = 1150mm Low-temperature side element: NF ℓ = 1150mm	High/low — NF-Single layer ℓ = 1140mm
Gas condition	Gas inlet: 280 ~ 300°C Gas outlet: 145 ~ 155°C	300 ~ 320°C 145 ~ 155°C
S.B. catalyst	6 ~ 7.5K saturated steam	6 ~ 7.5K saturated steam



Bag Filter

A Sumitomo bag filter is used with the high dust $DeNO_x$ process as the low temperature dust collector. This same bag filter was used successfully in a joint development project by Electric Power Development Co., Ltd., Sumitomo Heavy Industry Co., Ltd., and IHI. The results of this test confirmed the expected efficiency of the bag filter, as it succeeded in maintaining the concentration of dust below 10 mg/NM^3 with a pressure drop of 150 mm W.G. for the case where the flue gas had passed through the $DeNO_x$ process. A long-term test to determine the life span of the bag filter material is now in progress.

FGD PROCESS

Desulfurization Efficiency

When a boiler is fired with several kinds of coal, the SO_2 concentration of the flue gas may vary over a wide range.

In this test it was shown that a satisfactory desulfurization efficiency of over 96% can be obtained for SO_2 concentrations ranging from 300 to 1,000 ppm at the inlet.

Dust Removing Efficiency

When an FGD unit is installed as shown in Figure 1, the dust at the inlet is affected by the unreacted ammonium from the $DeNO_x$ process, dust contained in the flue gas, and other various gases such as fluorine, chlorine, etc.

According to the results of this series of tests, the effects of such dust and gases are relatively small. For instance, the concentration of dust at the outlet of the FGD system is less than $20/NM^3$, thus showing that this FGD system has sufficient dust removing capability.

Quality of Gypsum from the FGD Process

In the case of coal-fired boilers, traces of various impurities contained in the coal accumulate in the circulating scrubbing liquid of the FGD system after a period of time. This test showed that some of these impurities affect the gypsum crystallization and also control of pH in the scrubbing liquid. However, it was also shown that such problems can be solved and that gypsum of the desired quality can be obtained by taking appropriate measures such as injection of an additive. Furthermore, it was confirmed that an NH_3/SO_3 based product coming from the upstream side of the desulfurizing system does not significantly affect the quality of gypsum and that to obtain gypsum of high quality, the ash-separation method is more effective than the ash-mixed method.

Plugging of GGH Element

The GGH element is subject to plugging by mist deposits from the FGD process; but by operating the soot blower more frequently, this problem is not severe. During these tests, no significant corrosion or decrease in the thickness of the element wall was noticed.

FGD Booster Fan

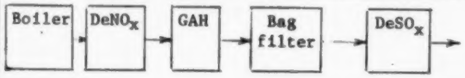
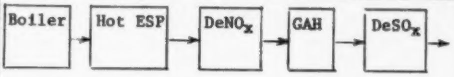
SUS material is used for the blades, and a rubber lined casing is used for corrosion protection. The booster fan has provided adequate and stable performance during these tests.

Waste Water Treatment System

Traces of various substances such as heavy metals, fluorine, chlorine, etc., contained in the coal and unreacted ammonium from the $DeNO_x$ process will accumulate in the circulating scrubbing liquid of the FGD system. Normally this liquid is blown down to protect the materials of construction from corrosion. This waste water stream is normally sent to an integrated waste water treatment system. A research program is now being conducted to develop a more economical waste water treatment system for FGD applications.

SECTION 7

EVALUATION OF TEST RESULTS AND CONSIDERATIONS FOR COMMERCIAL PLANTS

Item \ Evaluation		Evaluation of Test Results		Considerations for Commercial Plants
		High Dust Denitrifying Process	Low Dust Denitrifying Process	
Adaptability	Process			• In setting up the planning for actual plants, it is an important point to determine the process arrangement which depends on whether a low temperature or a high temperature dust collecting method is selected. The selection of the method largely affects the economy and general arrangement of the system and can be determined by considering the kind of coal used.
	Denitrification Function	• This process will not cause any serious plugging of the catalyst layer or the GAH and is useful from the viewpoint of the denitrification function. • As for wear, partial erosion was observed where there was a drift in the gas flow, but such phenomenon can be prevented by correcting the gas flow on the upstream side of the catalyst layer.	• In this process, the catalyst layer, especially the element of the GAH, is subject to more plugging compared with the high dust process, but this process has proved its effect in the DeNO$_x$ function if proper consideration is given to the construction of the catalyst layer and the form of the GAH element and if soot blow- is used appropriately.	• When applied to the actual system, unlike the case of the test plant, considerable changes will be expected in the gas flow, causing partial erosion of the catalyst at the upstream side. To minimize the erosion effect, the method of regulat- the gas flow must be considered carefully. • To solve the plugging problem that occurred in the low dust AH, it is necessary not only to select an appropriate value for the space velocity but also to follow the instructions given in the column at the left. Also, it is desirable to apply a close control on the NH_3/NO_x mol ratio to restrict unreacted NH_3 for partial load and to adopt a low conversion ratio for SO_3 for the catalyst.

SECTION 7 (cont'd.)

EVALUATION OF TEST RESULTS AND CONSIDERATIONS FOR COMMERCIAL PLANTS (cont'd.)

	Evaluation / Item	Evaluation of Test Results		Considerations for Commercial Plants
		High Dust Denitrifying Process	Low Dust Denitrifying Process	
Adaptability	Dust Collecting Function	• Unlike the low dust denitrification method, the NH_3-SO_3-based products from the GAH and ash containing NH_3 are collected almost completely by the dust collector installed on the downstream side. Therefore, the method of treating NH_3 ash becomes one of the problems to be considered. Thus, IHI is now conducting research on this problem in parallel with research on the effective use of NH_3 ash.	• A high temperature electrostatic precipitator is installed on the upstream side of the denitrification equipment, thereby freeing it from the effect of NH_3. This hot ESP is designed according to principles of conventional hot ESP design and performs well for a wide variety of coal and will not adversely affect the downstream side of the system. Since it does not contain NH_3, ash treatment is conventional.	• When using an ESP as the low temperature dust collector, its efficiency should be considered with the integrated system's dust collecting efficiency, including the dust removing efficiency of the wet type desulfurization equipment.
	Desulfurization Function	• Tests on the desulfurization function in conjunction with the high dust denitrification process have not been conducted yet; however, the NH_3-SO_3-based product and mist produced in GAH are collected almost completely by the dust collector on the downstream side of this process. Therefore, this process is more advantageous than the low dust denitrification process as far as the treatment of waste water and the removal of dust are concerned. Future testing will check treatment of waste water, integrated dust collecting efficiency, the requirement for a wet ESP, and the effect on the bag filter caused by the NH_3-SO_3-based product.	• When comparing the integrated flue gas treatment system with a conventional desulfurizing system without a $DeNO_x$ section, the problem of an NH_3-SO_3-based product arises from unreacted NH_3 entering the desulfurizing system and accumulating in the circulating liquid. This problem is solved by removing the ammonium through the waste water teatment system. Testing has shown that this process has an excellent desulfurizing efficiency and dust removing capability.	• The desulfurization process is conventional, but it is necessary to develop more economical techniques to treat chemical oxygen demand and to eliminate formation nitrogen from unreacted NH_3.

SECTION 8

CONCLUSION

This paper is a summary of the results of testing an integrated flue gas treatment system for a coal-fired boiler. Through these tests and associated research, it has been shown that the denitrification, dust collecting, and desulfurizing processes which have been tested are effective and that they can be incorporated into a full-scale integrated flue gas treatment system. These tests have provided the confidence that both the high and low dust $DeNO_x$ systems tested will be put into commercial use in the near future.

This test and research program will be continued in order to develop a more reliable integrated flue gas treatment system.

SECTION 9

APPENDIX

The Appendix Figures A-1, A-2, A-3, and A-4 show photographs of the denitrification, bag filter, and desulfurization test equipment, respectively.

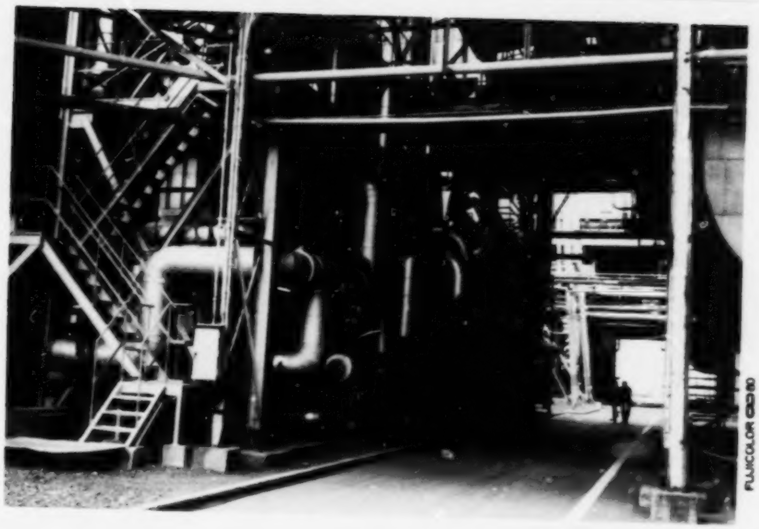


Figure A-1 Denitrification Test Equipment

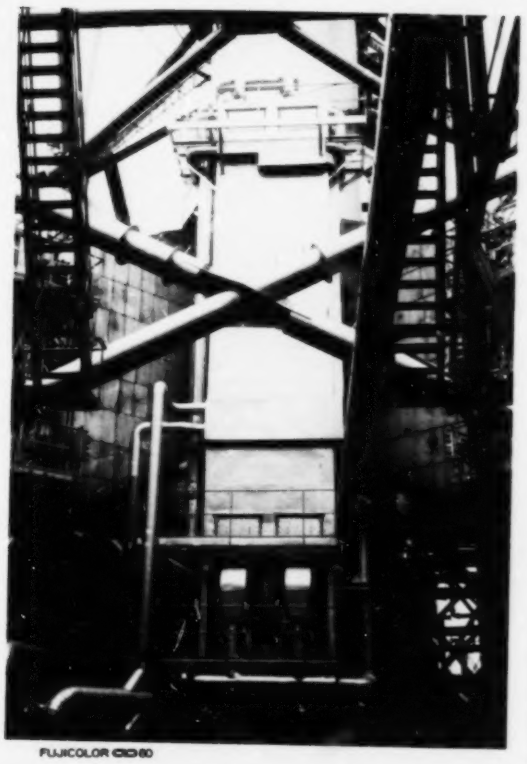


Figure A-2 Bag Filter

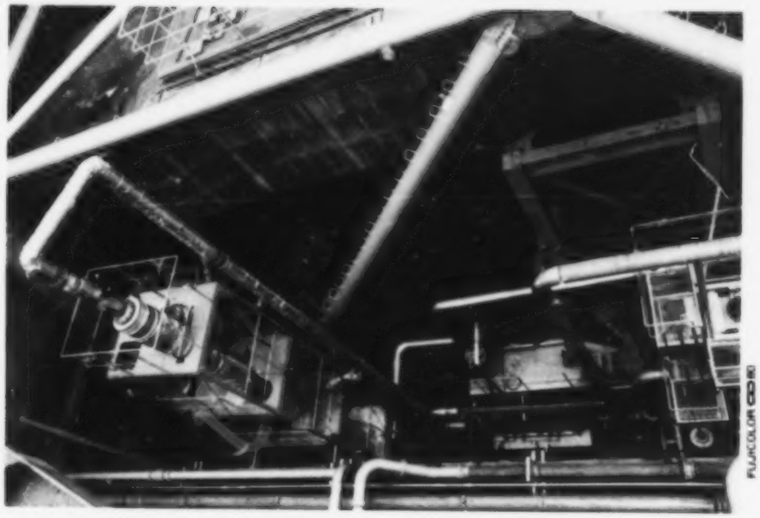


Figure A-3 Desulfurization Test Equipment

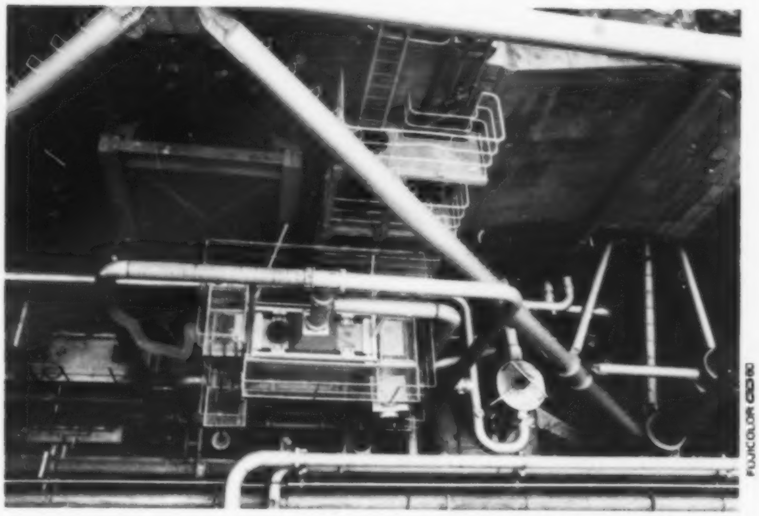


Figure A-4 Desulfurization Test Equipment

THE DEVELOPMENT OF A CATALYTIC NO_x REDUCTION SYSTEM FOR COAL-FIRED STEAM GENERATORS

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ABSTRACT

Work done recently by Mitsubishi Heavy Industries in Japan has resulted in the design and successful operation of a full scale catalytic NOx reduction system for coal-fired utility steam generators. This paper describes the program carried out to evaluate the commercial feasibility of catalytic nitrogen oxides removal from coal fired power plant flue gases. Also discussed is the design of a catalytic removal system for a large modern coal-fired central station.

Testing of pilot catalytic systems on coal fired steam generators was initiated at the Takasago station of EPDC in early 1977 using plate type catalysts. After two stages of testing, the plate type catalysts were replaced by grid type catalysts and testing was resumed (fall 1979). Since the conversion to grid type supports, more than 5000 hours of operation has been logged while maintaining more than 80 percent NOx removal in both low and high dust load environments.

At the Nakoso station of the Joban Joint Power Co. a grid type pilot catalytic system has been operating for over 10,000 hours with a removal efficiency of 84 percent. Operating under both high and low dust loads, draft losses (as at Takasago) have been maintained at low levels with only limited soot blowing during low dust loading and no soot blowing during high dust loading.

At the Shimonoseki station of the Chugoku Electric Power Co., the first full scale system for a coal-fired boiler (175-MW) in the world has run smoothly since startup in April of this year. Removal efficiency has been 51 percent, as expected, and the ammonia slip less than 1 ppm.

The paper also discusses ammonia slip, gas flow requirements, catalyst life and catalyst blinding from fly ash. The design of a 500-MW commercial unit based on the results of the test program is described and the various factors affecting large commercial design are discussed as well.

INTRODUCTION

In most nations, SOx, NOx and particulates contained in flue gas from steam generators must be minimized to meet environmental regulations. Particulates are normally removed by electrostatic precipitators or bag filters and SOx by tail-end flue gas treatment systems, but NOx removal is somewhat different. The NOx levels in flue gas have been reduced considerably by combustion modifications. However, there is a minimum level of NOx cencentration beyond which further reduction is not possible using these combustion modification techniques. By using a catalyst and a reductant (NH_3), however, the NOx generated in steam generators can be reduced by a process of dissocation to form N_2 and H_2O. This is known as the Selective Catalytic Reduction (SCR) process and is recognized as the most effective method for NOx reduction in Japan. The SCR process is performed at a gas temperature of about 300 to 400 C, which usually exists just upstream of the boiler air heater.

Since 1952, Mitsubishi Heavy Industries, Ltd. (MHI) has manufactured boilers for electric power plants under a technical license with Combustion Engineering, Inc. Almost all the boilers are tangentially fired. While combustion modification in tangentially fired steam generators has been adequate to meet the regulated NOx emission limits in the United States, the more stringent NOx emission regulations in Japan have led MHI to develop additional methods of NOx reduction: the very low NOx PM burner and the Selective Catalytic Reduction process.

To comply with increasingly stringent NOx regulations in the U. S. this MHI technology was licensed by C-E in April 1980 and will be available through C-E for steam generators in the U. S.

The very low NOx burner technology has been presented in another paper (1) at this conference, while this paper deals with MHI's SCR technology, focusing especially on its application to coal fired steam generators.

HISTORY OF DEVELOPMENT

MHI began developing a dry NOx removal process for boilers in 1971, concentrating efforts on the SCR process in 1973. The first application was for clean flue gas from liquefied natural gas (LNG) fired units, the second for semi-dirty flue gas from low sulfur oil fired boilers, and finally for dirty flue gas from high sulfur oil and coal fired boilers. Table I shows the SCR systems supplied by MHI.

As the clean flue gas from an LNG fired boiler does not contain particulate, it was possible to use a pellet type catalyst of 3 mm diameter packed in fixed bed reactors. A pilot plant with a capacity of 10,000 Nm^3/h^* was constructed at a 350-MW boiler at the Tokyo Electric Power Co's, Minami-Yokohama power station. This unit demonstrated a 10,000-hr catalyst life by the end of 1975, without any performance deterioration (2). This successful test was MHI's first experience with the SCR process and it provided the basic technical information for further development. The results of this test were applied in the first, large full-scale system at the newly installed 600-MW unit at Kyushu Electric Power Co's, Shinkokura station. It began operation in June 1978 and has operated successfully since then.

In applying the process to the semi-dirty light oil firing flue gas, an intermittent moving bed reactor was first developed using a pellet type catalyst, but it was later concluded that fixed bed reactors using parallel passage catalysts, such as plate or grid types, would be more suitable and practical for both operation and maintenance. MHI carried out an evaluation program for both types of catalyst simultaneously and, after considerable studies, concluded that the grid catalyst is more economical for both oil and coal firing, primarily because of the compactness of the reactor chamber.

Grid shaped catalysts used in parallel passage reactors are manufactured by extrusion molding. Two types are commercially available; one is a homogeneous type, where the catalyst is formed by a uniform blend of active and support material, the other is a coated type, in which the active catalyst material is coated on a ceramic support structure (substrate). Either type of catalyst may be mass-produced through a series of processes to be within a predetermined tolerance of dimension, strength and activation performance. Figure 1 shows one element of the grid type catalyst.

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*Normal cubic meters per hour

The first commercial system using a grid catalyst was for an oil-fired boiler at the Sodegaura Refinery of the Fuji Oil Co., a 160 t/h boiler (Fig. 2) put into operation in December 1977 (3). This unit has operated smoothly for more than two years without the replacement of the original catalyst. In January 1980 a 700 MW oil-fired unit, Chita 4, owned by the Chubu Electric Power Co., was put into service. It is the largest SCR application to an oil-fired boiler in the world.

To develop the SCR system for coal fired boilers, MHI constructed a pilot plant known as the "DM-600 project" on a coal-fired boiler at the Takasago Power Station of the Electric Power Development Co. (EPDC) and put it into operation in January 1977 as a joint development project with EPDC. In March 1979, the second pilot plant was put into operation at Nakoso Power Station of the Joban Joint Electric Power Co. sponsored by Tokyo Electric Power Co., Tohoku Electric Power Co., and Joban Joint Electric Power Co. and called the TTJM project. This pilot plant consists of a precipitator to be usable either in the hot or cold temperature mode, two SCR systems, one for high dust and another for low dust loads, a wet desulfurization scrubber, and water treatment systems. The purpose of this project was to investigate the overall performance of the total pollution control package as well as individual systems. This configuration permitted testing with the low dust conditions in the case of the hot electrostatic precipitator upstream of the SCR reactor and with high dust conditions in the case of the low temperature electrostatic precipitator downstream of the air heater.

The test results of the TTJM project were used in designing the first SCR system for a full-scale coal-fired commercial unit, the Shimonoseki 1 boiler owned by the Chugoku Electric Power Co. This is a 175-MW unit with a high concentration of fly ash. It was put into operation in April 1980 and has operated trouble free.

These projects are summarized in Fig. 3 and the test results are described in a later section of this paper.

DEVELOPMENT OF CATALYST

FEATURES

The catalyst for the SCR process is made of a mixture of active material and support material. The active material has to be highly active, durable, and formed to have sufficient surface and porosity to be efficient. In addition, it must not adversely affect the boiler. Both materials must be low in captial and replacement costs. The overall structure must be strong, compact, and low in draft loss. A special consideration is that in most chemical processes, temperature and pressure can be controlled for the most suitable reaction conditions for the catalyst, but in flue gas treatment, the catalyst must be developed to meet the boiler's operating conditions.

PROCEDURE TO DEVELOP THE CATALYST

MHI's policy regarding the production of catalysts for SCR plants is to develop them with well established catalyst manufacturers who then manufacture them in accordance with MHI specifications. To do this, MHI established a total evaluation program of the catalyst development including trial formulation, evaluation of chemical activity and physical properties, and testing for durability. This is illustrated in Fig. 4.

The first step in developing a catalyst is to select its composition and determine the preparation method, then, with the catalyst manufacturer's cooperation, to establish the specification after evaluation of bench scale testing program carried out at MHI's Hiroshima Technical Institute.

The catalyst manufacturers provide MHI with catalysts suitable for each specific application. MHI carries out pilot tests to confirm the catalysts' durability in actual flue gas. These tests consist of accelerated deterioration tests, such as alkaline dust heating to predict the life expectancy of various fundamental tests, such as adhesion, clogging, and abrasion with different kinds of dust, and testing the rejuvenation response of deteriorated catalyst. Moreover, MHI is making a considerable effort to find the causes of catalyst deterioration by evaluating various results of analyses such as X-ray diffraction, X-ray fluorescence, X-ray micro probe analysis, X-ray photoelectron spectroscopy, and mass spectrometry. Physical properties are tested, such as surface, area, pore volume, and strength against crushing, bending, abrasion, thermal shock, etc.

CONDITIONS FOR SELECTING CATALYSTS

There are large differences in SOx and dust concentration among exhaust gases from various fuels. It is essential to select the most suitable catalyst for each specific exhaust gas. The basic approach to the selection of the catalyst for use with coal fired boiler exhaust gas is summarized as follows.

RESISTANCE TO SOx

Since coal-fired boiler exhaust gas normally contains high concentration of sulfur oxides, the catalyst should be tolerant to SO_2, the formation of SO_3 by the catalyst becomes a particularly important problem because of its detrimental effect on the equipment downstream. A catalyst should be selected that is capable of minimizing the SO_2 oxidization rate. Figure 5 shows a comparison between restrained and non-restrained catalysts in the formation of SO_3.

WORKING TEMPERATURE

With an exhaust gas containing NH_3 and SO_3, the catalyst can deteriorate from a low gas temperature. The boiler may have to be modified so that the NOx removal plant can be continuously operated at a temperature above the allowable minimum working temperature of the catalyst. These minimum working temperatures for various SO_3 concentrations are known for operational results obtained in the past.

LONG LIFE

It is necessary to select a catalyst that is stable thermally and stable with regard to alkali compounds (K_2O in particular), contained in dust that can cause deterioration. As shown in Fig. 6, according to the forced deterioration test using the alkali compounds as a pollutant, a catalyst of MHI specification, which consists of TiO_2 as the support material and a proprietary blend of two other active materials X and Y shows a much more stable performance than normal catalyst of a single active component.

FREEDOM FROM DUST DEPOSITION

It is necessary to select a catalyst with a structure (pitch) that will not become plugged by dust contained in exhaust gas. Experience with catalysts with 7 to 10 mm nominal pitch have shown satisfactory results even with high concentration of dust.

SOOT BLOWING RESISTANCE

If dust should adhere to the catalyst, soot blowing is the most effective way to remove it. The catalyst must be strong enough to withstand this soot blowing. Figure 7 shows a comparison between our improved erosion resistant type and a common catalyst.

RESISTANCE TO EROSION

Erosion of the catalyst from fly ash in exhaust gases cause a decrease in the life of the catalyst. It is necessary to give special consideration to this, especially when exhaust gases contain a high concentration of dust. Without any counter measures for the catalyst, the leading ends of catalyst can be severely eroded. MHI uses a catalyst with a high erosion-resistance made by a propietary method. In addition to this, a section of dummy catalyst support located upstream of the active catalyst can be used to absorb the end impact and act as guide vanes, thus protecting the active catalyst downstream. If the above measures are used and the appropriate gas velocity provided, the erosion of the catalyst can be eliminated. Figure 8 shows the test results comparing erosion resistant catalyst with common types.

OUTLINE OF RESULTS IN FIELD TESTS

The operation of three coal-fired test plants as shown in Fig. 3 has provided us with extensive information in establishing the design philosophy of NOx removal systems. We can summarize these operating results as follows:

DM-600 Project

This pilot plant project was initiated in January 1977, as a joint research program between MHI and EPDC for the purpose of establishing the selection and durability criteria for catalysts for coal fired boilers. A flue gas slipstream was taken from the 250-MW No. 1 coal-fired unit at the Takasago power station of EPDC. In the first and second stages of this program, the plate type catalyst developed by MHI was tested and it was proven that parallel flow, fixed bed type reactors would work very will for either high or low ash laden gas. These two stages of the program involved continuous operation of more than about 10,000 hours.

As the third stage of the program, a grid type catalyst with 10 mm pitch square holes was tested beginning in August of 1979. The composition of the flue gas at the inlet of the reactor for the high dust system included ∿12 to 15 g/Nm^3 of dust, about 1500 ppm of SO_x and about 250 ppm of NO_x. Analysis of fly ash showed about 48% SiO_2 and 23% Al_2O_3.

As shown in Fig. 9, it was confirmed that the NOx removal efficiency could be maintained at more than 84% with less than 3 ppm of ammonia slip at NH_3/NO_x ratio 0.85. The draft loss across the reactor did not change throughout the test period for the high dust system. For the low dust system, where the dust loading was about 50 mg/Nm^3, there was a slight increase in draft loss across the reactor after 1000 hours operation, and it reached a 40mm H_2O increase after 3000 hours operation. But after soot blowing, the draft loss increase was completely eliminated. Except for this, the low dust system operation showed almost the same performance as the high dust system.

These tests are being continued until the end of 1980.

Prior to pilot plant operation it was feared that alkaline compounds especially those of sodium and potassium could impair actibity of the catalyst by chemical reaction with active catalyst materials. Such alkaline compounds in fly ash are known to form a glassy complex of Al_2O_3-CaO-Na_2O-K_2O in the liquid phase and become non-active when cooled. After laboratory inspection of catalysts taken from the pilot plant, we found no accumulation of alkaline compounds in the catalysts, indicating that alkaline poisoning of the catalysts can be eliminated.

According to laboratory experiments, the rate of catalyst erosion increases rapidly when the gas velocity is greater than 10 meters/sec or when the particulate size is over 40 microns in diameter. In the case of the high dust system, while the average diameter of fly ash particle is about 20 microns, considerable particles of more than 40 microns are present. This results in an erosive condition. In a test with the gas flow rectified by an upstream dummy layer of catalyst support material and the gas velocity below 10 m/s, we eliminated the erosion of catalysts.

The smaller the particle size of fly ash and the more alkaline compounds it contains, the more adhesive the fly ash will be. In the case where the SCR reactor was located downstream of the hot precipitator, the dust particles that escaped capture were mostly 3 microns or less in size. This material proved to be somewhat sticky therefore soot blowers have had to be operated periodically to remove this fine fly ash that collected on the surface of catalysts.

TTJM Project

This project was initiated to establish an integrated flue gas treatment system for coal-fired power plants. The pilot plant was installed at the Nakoso power station of Joban Joint Power Co., Ltd. This boiler used mixed combustion of coal and heavy oil (Coal amounting to about 40 percent during the day time and to about 70 to 80 percent during the low load period at night.) As shown in Fig. 3, the pilot plant consists of not only high and low dust NOx removal equipment, but also includes wet desulfurizing equipment but also includes wet desulfurizing equipment and waste water treatment equipment so that the overall performance of the integrated clean-up system in various combinations could be thoroughly tested. Testing was started in

April 1979. At full boiler load, the flue gas in the high dust system contains about 7 grams/Nm^3 of dust, about 240 ppm of SOx and about 300 ppm of NOx with fly ash composition having about 55% SiO_2 and 27% Al_2O_3. As shown in Fig. 10 with a vertical reactor chamber using a grid catalyst with an Sv of 2200 H^{-1} NOx removal efficiency was maintained at more than 84% and less than 2 ppm of ammonia slip with NH_3/NOx ratio of 0.85 after about 10,000 hours operation. The draft loss across the reactor chamber did not increase during the period of high dust load operation. In the low dust system (100-300 mg/Nm^3 dust loading) the draft loss showed a gradual increase and reached about 60 mm H_2O increase after 3000 hours operation. At this point, the draft loss was successfully reduced to the initial value through soot blower operation.

The regenerative air heater in the high dust system has the normal arrangement of heating elements, DU type for high and middle temperature zones and NF type for the low temperature zone. Air heater plugging due to the deposit of ammonia bi-sulfate did not occur. This was attributed to the sand blasting effect of highly concentrated fly ash and operation of the soot blowers once a day. In the low dust system, the arrangement of air heating elements was revised to use the SNF type (3.5 mm pitch) for the middle and low temperature zones because draft loss increase across the airheater had been observed with original airheater. With this modification and by operating soot blowers both up and dwonstream three times per day, continuous pluggage-free operation was attained. This is shown in Figs. 11 and 12.

FULL SCALE DEMONSTRATION

The Shimonoseki Unit of Chugoku Electric Power Co. is a 175-MW coal-fired boiler and was constructed in 1967. The NOx removal equipment was installed in 1980 with grid catalysts having 10-mm pitch square holes. The reactor chamber is a vertical down flow unit and is located at the side of air heater and precipitator. High dust laden flue gas is taken from the economizer outlet and directed to the top inlet of the reactor and returned to the inlet of the air heater. A provision has been made to keep the inlet gas temperature above 330°C at lower loads by gas bypassing of the economizer.

The system began operation in April, 1980 as the first full scale coal fired SCR system in the world. The plant has been running since then without any trouble at above design NOx removal efficiency, and ammonia slip concentration less than 1 ppm. The design removal target for this system was at least 50 percent. Design conditions are shown in Table II and plant layout in Fig. 13.

The initial test results for NOx removal efficiency and ammonia slip concentration at various boiler loads are shown in Fig. 14. This data was taken after one month operation, when the catalyst was in a fresh condition. However, we are sure that deterioration of catalyst activity will not occur at least within one year operation, judging from our test results at other pilot plants.

NO_X REMOVAL SYSTEM FOR 500-MW BOILER

OUTLINE OF DESIGN

The following outlines the design of a NOx removal system to be applied to a new 500-MW coal-fired steam generator. The flue gas capacity is 1,553,000 Nm^3/h at 4.3% O_2 when firing a typical sub-bituminous coal at the maximum continuous rating of the boiler. Design conditions for two cases, NOx removal efficiency of 80% (Case I) and 90% (Case II) are shown in Table IV.

MHI has decided to use a 7 mm pitched catalyst for high dust systems on coal fired boilers since it was proven that dust clogging would not occur on the TTJM project. In this instance the reactor is located between economizer and air heater and high ash-laden gas will be treated.

The specification of the equipment is shown in Table IV. The reactor chamber is a vertical type where flue gas enters the chamber from the top and passes downward through three or four stages of catalyst (Case I & II respectively). Each catalyst layer consists of a group of catalyst modules having a cross-sectional dimension of about 2 x 1 meters a height of 1.65 meters, the weight of a single module is about 3 tons and 216 or 288 modules are required for Cases I & II respectively. Each catalyst module can be carried in and out of the reactor chamber using an electric traveling hoist installed within the steel structures of reactor chamber. One element of the catalyst is 150 x 150 mm square, about 650 millimeters in length, and about 14 kg in weight. A total of 25.920 elements are required in Case I and 34,560 elements in Case II.

Eighteen traveling frame soot blowers in Case I or 24 sets in Case II using steam as a blowing medium are installed for cleaning the catalysts. Low pressure, orifice-type ammonia injection nozzles are installed upstream of the reactor chamber, followed by an ammonia/flue gas mixing device. The plant layout for Case I is shown in Fig. 15 and Figure 15 shows a schematic flow diagram of the system. The predicted NOx removal efficiency and ammonia slip concentration curves are shown compared to boiler load in Fig. 17 for both cases, and Fig. 18 shows the relationship between ammonia to NOx ratio and NOx removal.

PROCESS DESIGN CONSIDERATIONS

There are three types of fixed bed catalyst reactor chambers where grid catalyst elements are used: vertical up-flow, vertical down-flow and horizontal flow. The vertical down-flow type is generally preferred in SCR systems for coal fired boilers because it offers the following advantages:

(1) Less dust deposition on the catalyst surfaces and less plugging risk than the horizontal flow type reactor chamber.

(2) Less dust deposition on the tope ends of the catalyst element than the gas up-flow type reactor chamber.

There are two major factors to be carefully considered when the gas velocity through the system is determined; one is the erosion of catalyst elements and the other is the dust deposition on the catalyst during low load operation with reduced gas velocity. The following are the results observed in the pilot plant tests:

(1) There is no accumulation of dust deposits on the ends of catalyst layers in vertical flow reactors when the gas superficial velocity is 2 m/sec or greater. (Even on the inside surfaces of catalyst there is no growth of dust deposit with such a low gas velocity.)

(2) A low gas velocity is desirable for minimizing erosion of the catalyst elements. However, the catalyst elements developed by MHI can be used without trouble with dust laden gas flows of up to 10 m/sec.

In designing a NOx removal system, therefore, the gas velocities are selected taking the anticipated minimum and maximum load of the plant into careful consideration. In the case of heavily dust-laden flue gas, an erosion resistant dummy layer should be provided ahead of the catalyst layers to rectify the stream of gas and dust.

If the flue gas temperature becomes too low, the reaction products of SO_3 and HH_3 are formed on the active surfaces of the catalyst, impairing the capability of the catalyst. However, should the flue gas temperature return to a high level the deposits can be carried away by the gas stream, restoring the capability of the catalyst. The degree of reduction in NOx removal capability and the period of time required for its restoration largely depend upon the gas temperature and the SO_3 concentration of the flue gas. Thus, the operating temperature range of the catalyst should be determined to be suitable for the flue gas condition.

When the flue gas has a high SO_3 concentration, there is a possibility of corrosion and plugging in the air preheater due to the ammonia-sulfur compound. The dust in the stack gas will also be adversely affected by SO_3 leaving the air preheater. To avoid such difficulties, the SO_2 to SO_3 conversion due to the catalyst should be controlled to a minimum. Therefore, the catalyst should be controlled to a minimum. Therefore, the catalyst used in the NOx removal systems should be tested to meet a maximum rate of conversion criterion of SO_2 to SO_3 of about 1% when treating a 1500 ppm SO_2 concentration flue gas.

STRUCTURAL DESIGN

Figure 19 shows a cut-away view of the vertical type reactor chamber. As the reactor chamber is a massive high temperature structure, its supporting points at four corners are allowed to slide to permit thermal expansion of the structure. The reactor chamber is fixed only at a point in the middle of its front wall. The midpoints of the other three walls are provided with guide stoppers through which the horizontal forces exerted to the chamber are conveyed to the supporting steel.

A number of catalyst elements are packed into steel grids to form catalyst modules and these catalyst modules are placed on a rack having several shelves. The gaps between modules are sealed to prevent short circuiting of the gas; attention is also paid to prevent ash deposits. The racks are fabricated from steel and supported at four bottom corners by the main frame, which forms a part of the boiler house supporting steel structure. The catalyst modules may be moved in and out of the reactor chamber through openings on the side of the chamber by removing cover plates. To facilitate handling of the catalyst modules, platforms are provided at the heights of the respective module shelves. A hoist is provided to lift the modules to and from these platforms. Soot blowers are provided for each shelf of catalyst modules and located at a right angle to the direction of module removal and replacement.

The ammonia distribution and mixing devices are located immediately upstream of the catalyst. These are constructed from mild steel pipe and structural members to form a grid arrangement which assures thorough mixing of ammonia with the entering flue gas.

CONCLUSION

To meet the increasingly stringent NOx emission regulations in highly industrialized countries, a combination of combustion modification and flue gas treatment will soon be necessary. To that end the development of the Selective Catalytic Reduction system for boiler application has been successfully demonstrated in full scale commercial units for all fossil fuels.

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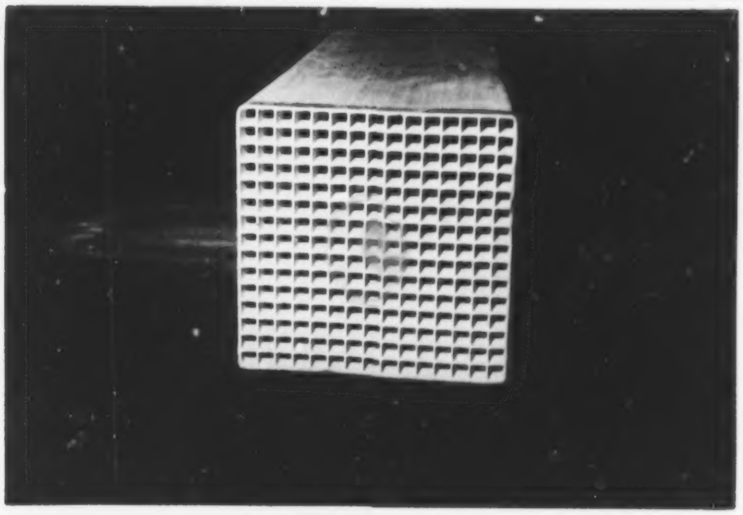

Fig. 1: Grid-type catalyst

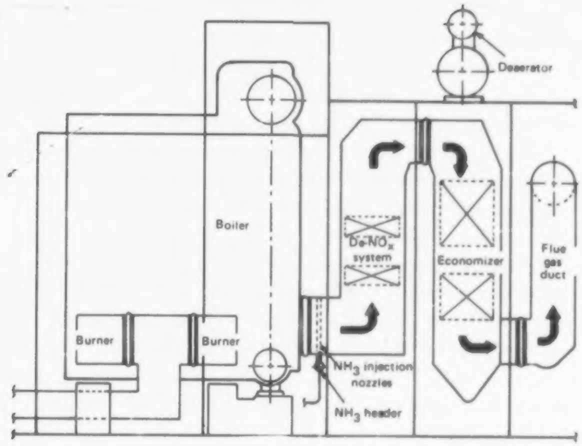
Deaerator
Boiler
De-NO$_x$ system
Economizer
Flue gas duct
Burner
Burner
NH$_3$ injection nozzles
NH$_3$ header

Fig. 2: General arrangement of 160 t/h oil-fired boiler at the Sodegaura Refinery of the Fuji Oil Co.

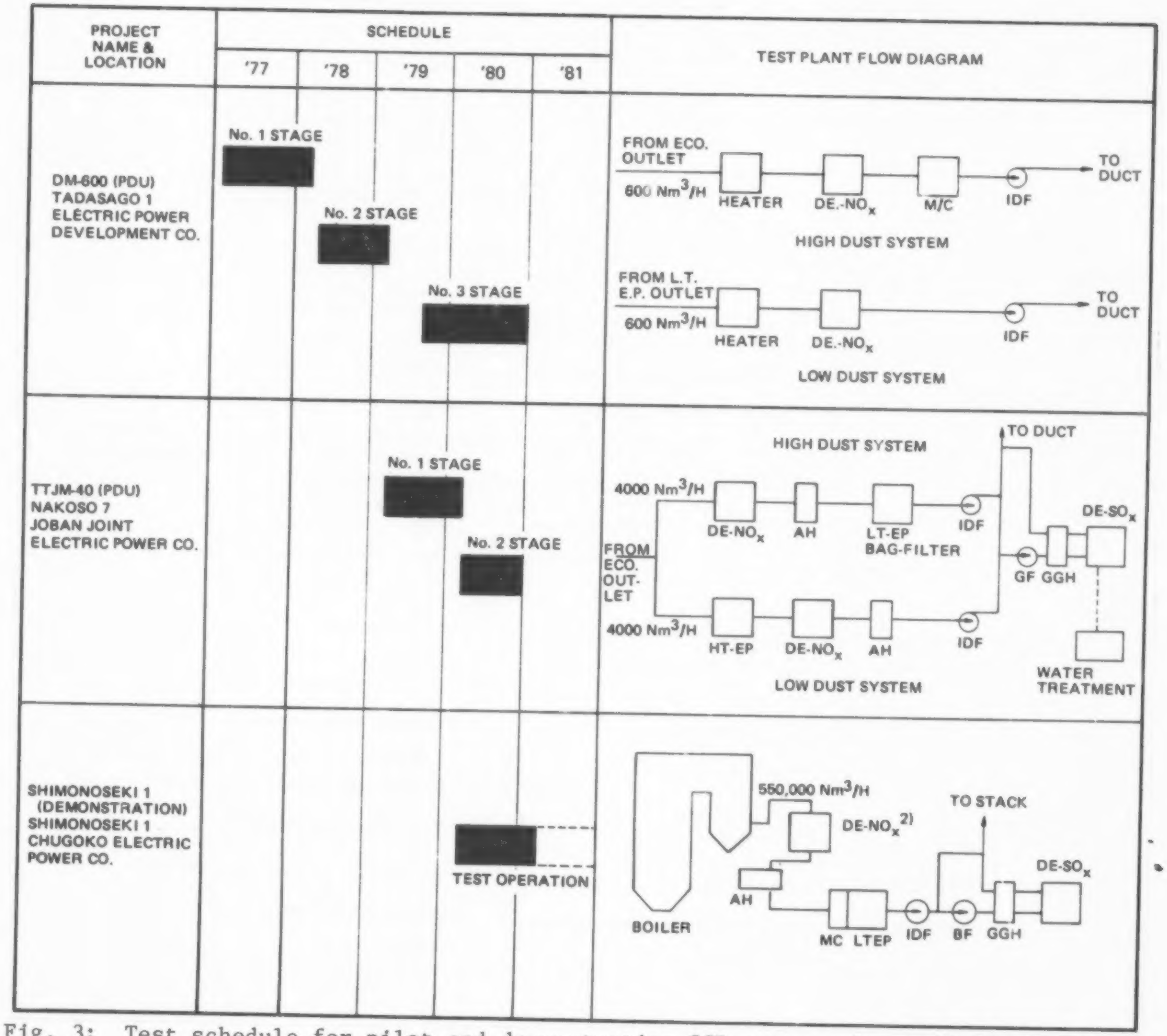
PROJECT NAME & LOCATION
SCHEDULE
'77
'78
'79
'80
'81
TEST PLANT FLOW DIAGRAM
DM-600 (PDU)
TADASAGO 1
ELECTRIC POWER DEVELOPMENT CO.
No. 1 STAGE
No. 2 STAGE
No. 3 STAGE
FROM ECO. OUTLET
600 Nm³/H
HEATER
DE.-NOx
M/C
IDF
TO DUCT
HIGH DUST SYSTEM
FROM L.T. E.P. OUTLET
600 Nm³/H
HEATER
DE.-NOx
IDF
TO DUCT
LOW DUST SYSTEM
TTJM-40 (PDU)
NAKOSO 7
JOBAN JOINT
ELECTRIC POWER CO.
No. 1 STAGE
No. 2 STAGE
HIGH DUST SYSTEM
TO DUCT
4000 Nm³/H
DE-NOx
AH
LT-EP BAG-FILTER
IDF
DE-SOx
FROM ECO. OUT-LET
GF
GGH
4000 Nm³/H
HT-EP
DE-NOx
AH
IDF
WATER TREATMENT
LOW DUST SYSTEM
SHIMONOSEKI 1
(DEMONSTRATION)
SHIMONOSEKI 1
CHUGOKO ELECTRIC POWER CO.
TEST OPERATION
550,000 Nm³/H
DE-NOx 2)
TO STACK
DE-SOx
AH
BOILER
MC
LTEP
IDF
BF
GGH

Fig. 3: Test schedule for pilot and demonstration SCR systems for coal-fired boilers

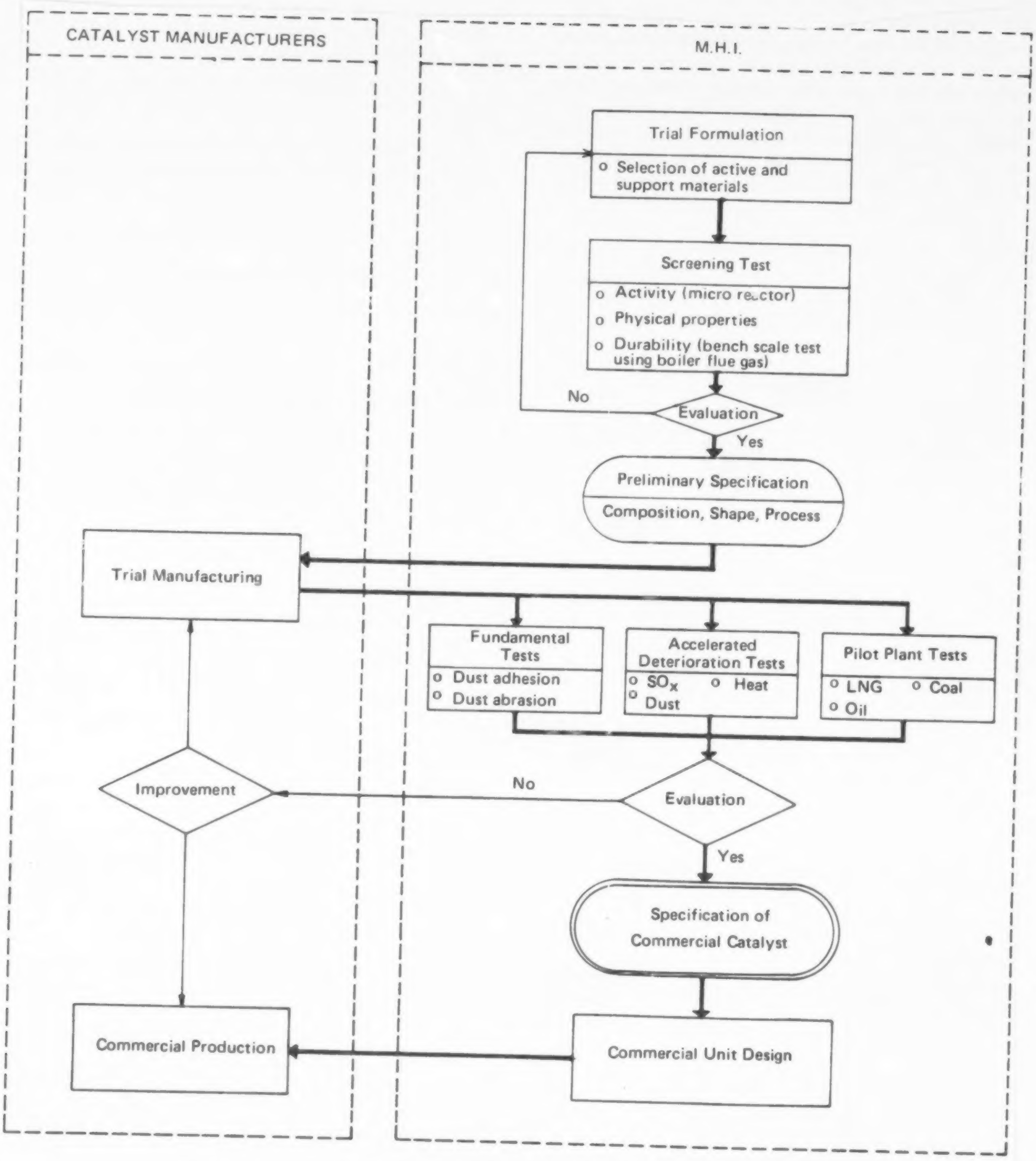
CATALYST MANUFACTURERS
M.H.I.
Trial Formulation
o Selection of active and support materials
Screening Test
o Activity (micro reactor)
o Physical properties
o Durability (bench scale test using boiler flue gas)
No
Evaluation
Yes
Preliminary Specification
Composition, Shape, Process
Trial Manufacturing
Fundamental Tests
o Dust adhesion
o Dust abrasion
Accelerated Deterioration Tests
o SOx
o Heat
o Dust
Pilot Plant Tests
o LNG
o Coal
o Oil
Improvement
No
Evaluation
Yes
Specification of Commercial Catalyst
Commercial Production
Commercial Unit Design

Fig. 4: Development of NOx reduction catalyst

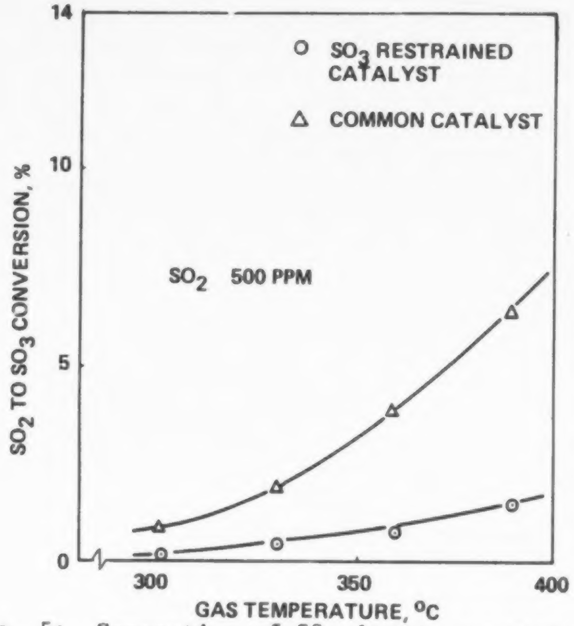

Fig. 5: Generation of SO_3 due to catalyst

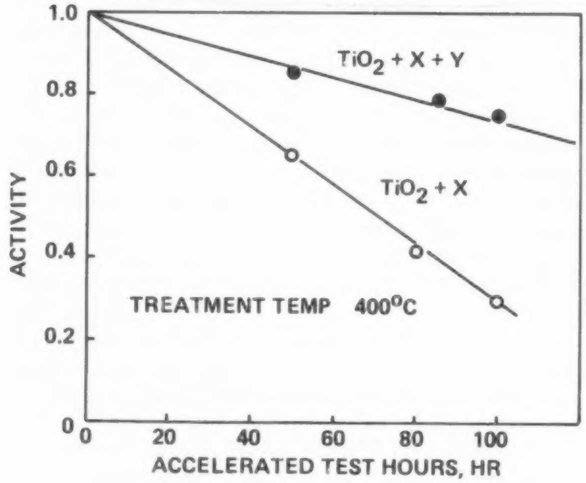

Fig. 6: Test results of accelerated deterioration test with alkalimetal sulfate

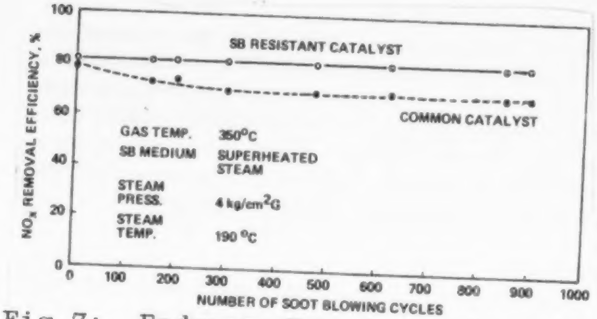

Fig.7: Endurance test results with soot blowing

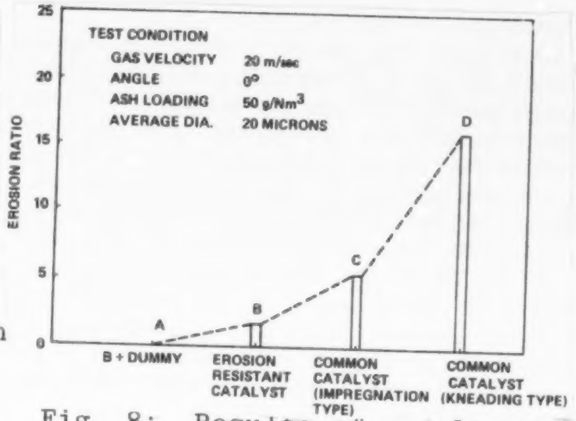

Fig. 8: Results of accelerated erosion test

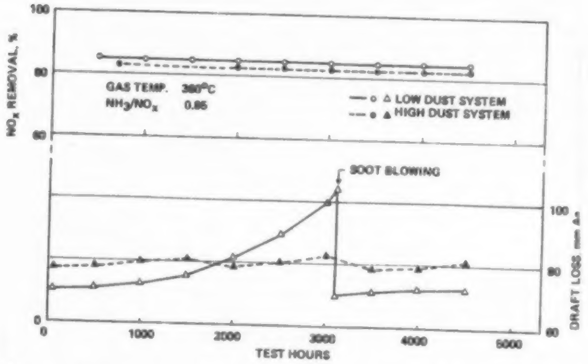

Fig. 9: Test results of pilot plant at DM-600 project

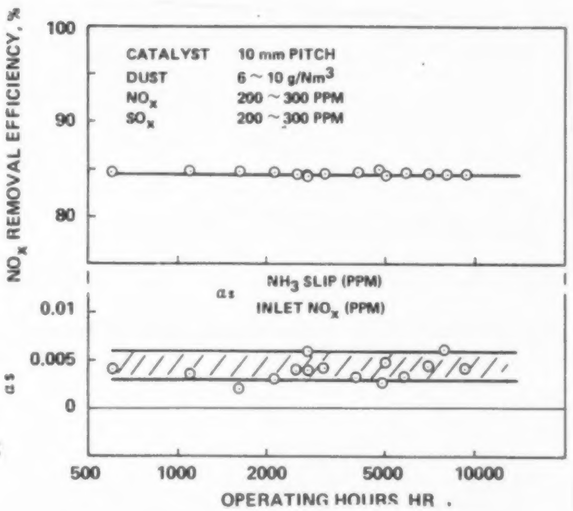

Fig. 10: Test results of TTJM project

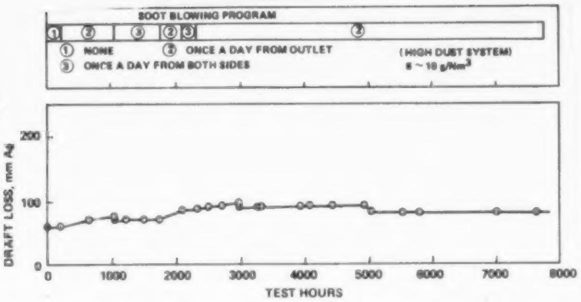

Fig. 11: Results of air heater air side draft loss versus soot blowing at TTJM

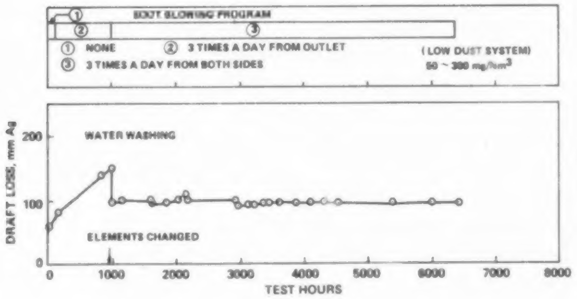

Fig. 12: Results of air heater air side draft loss versus soot blowing at TTJM

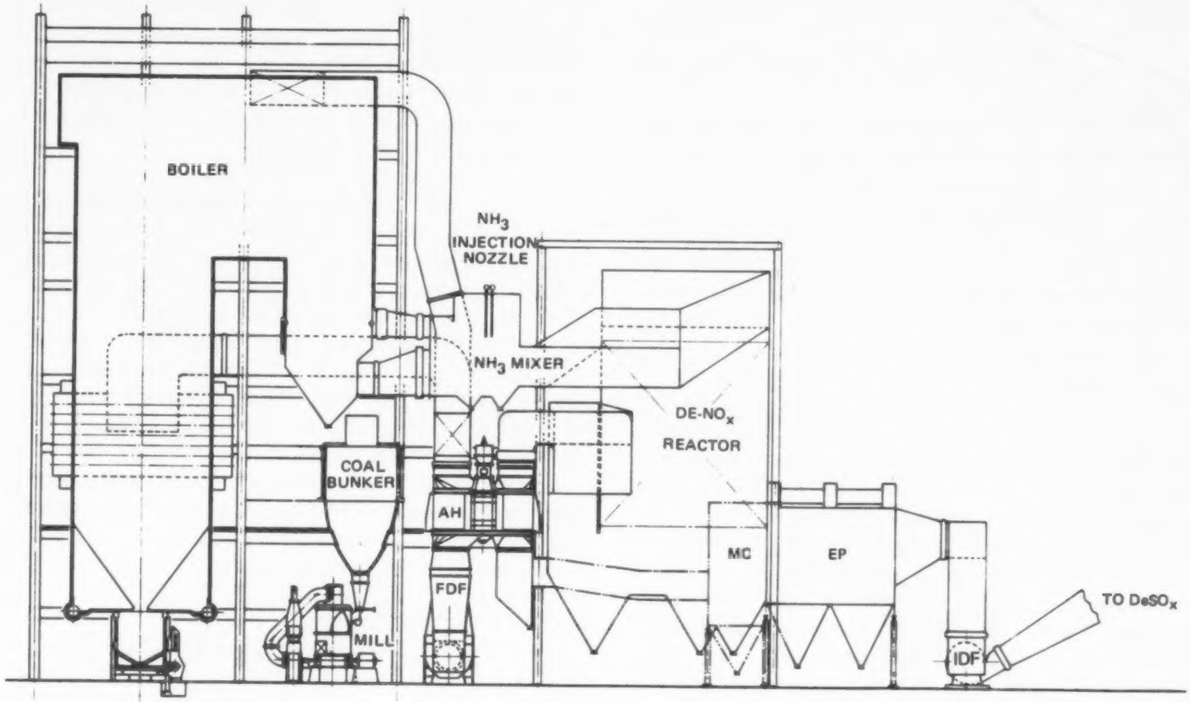
BOILER
NH_3 INJECTION NOZZLE
NH_3 MIXER
COAL BUNKER
AH
FDF
MILL
DE-NO_x REACTOR
MC
EP
TO $DeSO_x$
IDF

Fig. 13: General arrangement at Shimonoseki No. 1 boiler

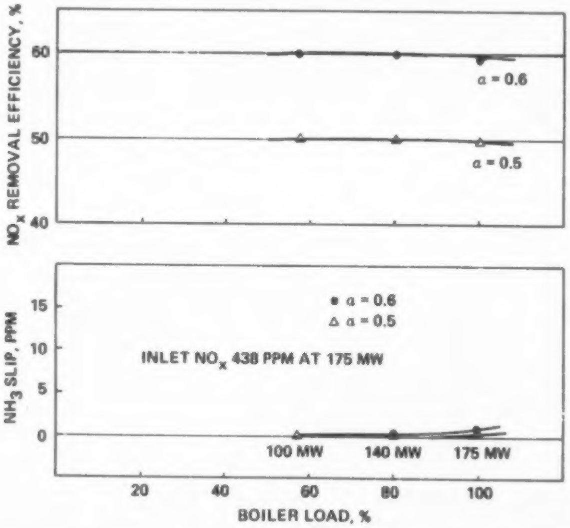

Fig. 14: Initial performance test results at Shimonoseki No. 1 unit

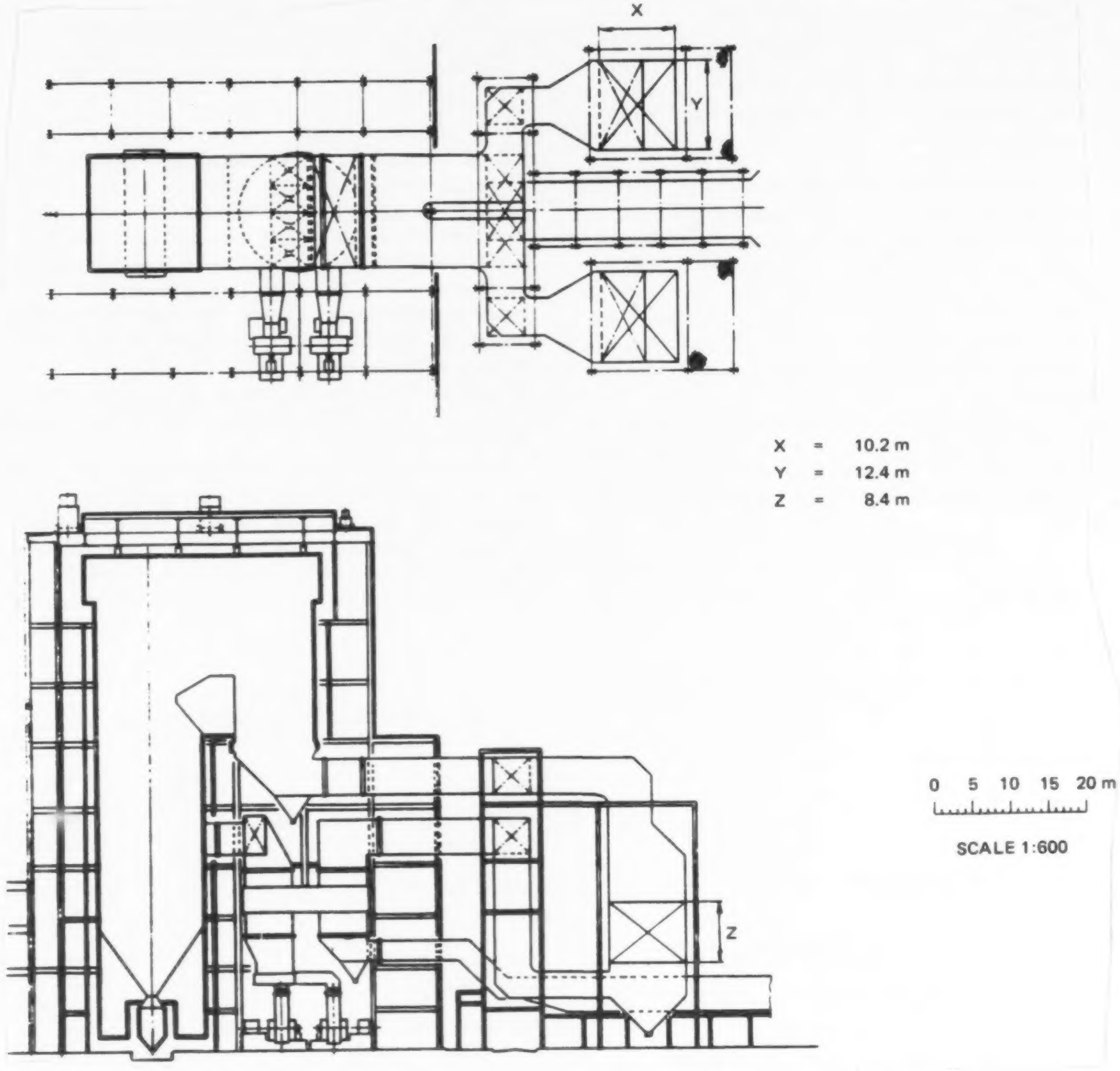
X
Y
X = 10.2 m
Y = 12.4 m
Z = 8.4 m
Z
0 5 10 15 20 m
SCALE 1:600

Fig. 15: General arrangement of 500 MW coal-fired boiler with SCR system (Case I)

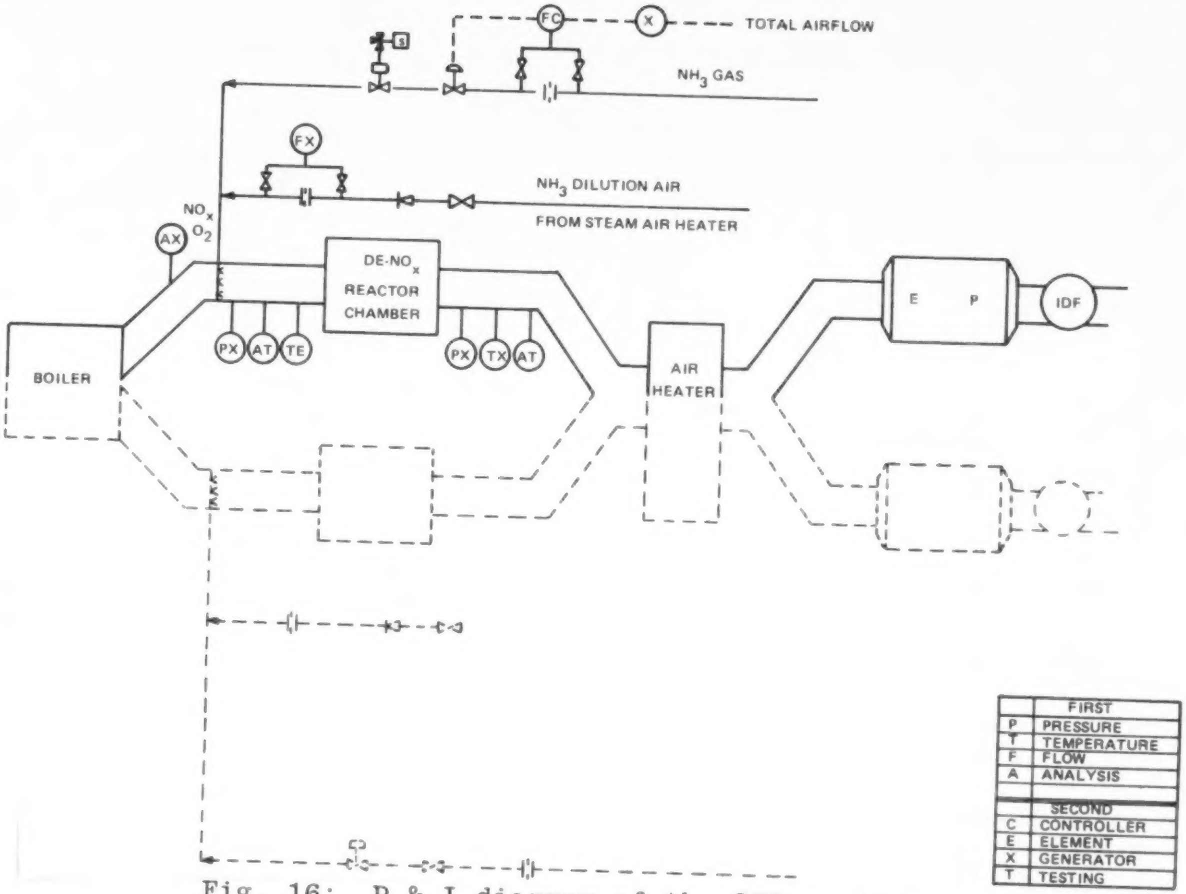
TOTAL AIRFLOW
FC
X
S
NH_3 GAS
FX
NH_3 DILUTION AIR
FROM STEAM AIR HEATER
NO_x
O_2
AX
DE-NO_x
REACTOR
CHAMBER
BOILER
PX
AT
TE
PX
TX
AT
AIR
HEATER
E
P
IDF
FIRST
P PRESSURE
T TEMPERATURE
F FLOW
A ANALYSIS
SECOND
C CONTROLLER
E ELEMENT
X GENERATOR
T TESTING

Fig. 16: P & I diagram of the SCR system

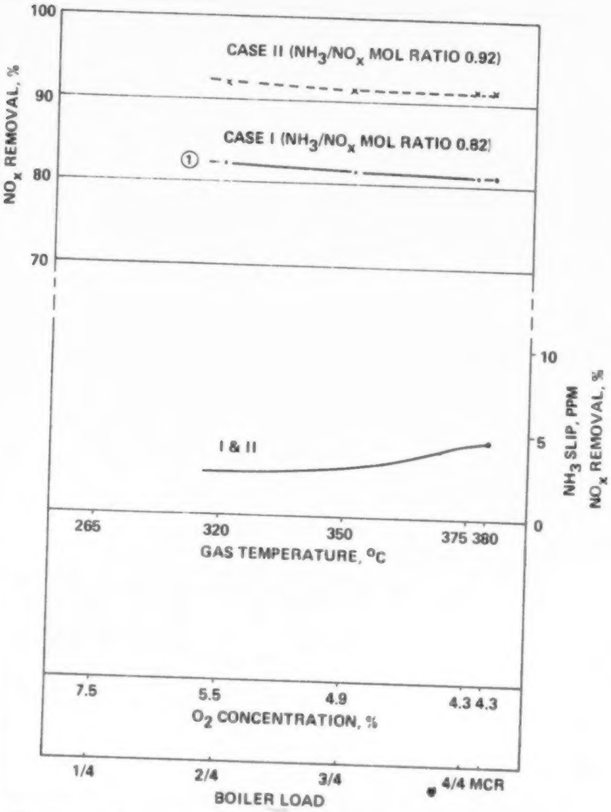

Fig. 17: Predicted NOx removal performance versus boiler load

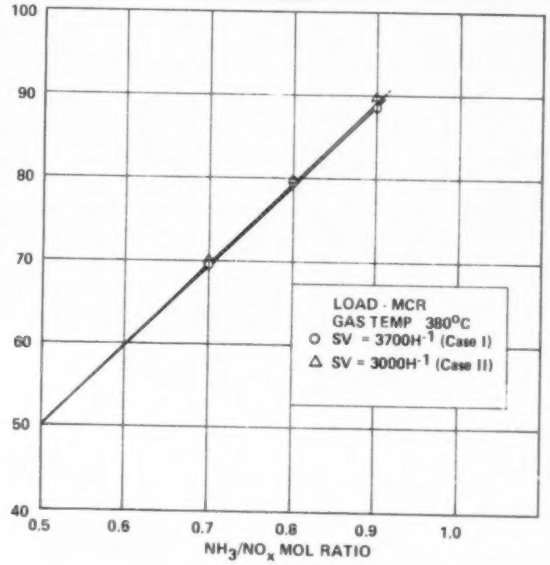

Fig. 18: Predicted NOx efficiency vs NH_3/NOx mol ratio

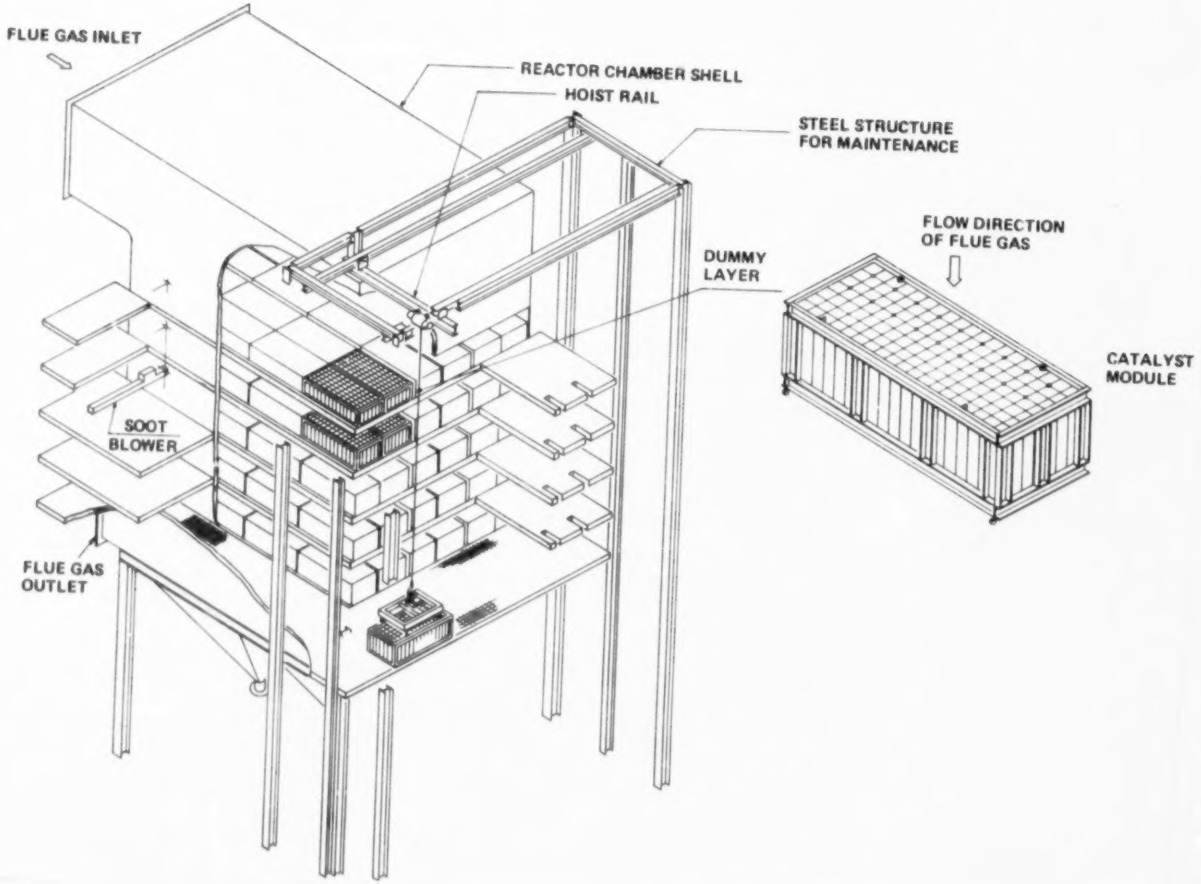
FLUE GAS INLET
REACTOR CHAMBER SHELL
HOIST RAIL
STEEL STRUCTURE FOR MAINTENANCE
FLOW DIRECTION OF FLUE GAS
DUMMY LAYER
CATALYST MODULE
SOOT BLOWER
FLUE GAS OUTLET

Fig. 19: Cutaway view of Reactor Chamber

TABLE I
SUPPLY LIST OF MHI SCR SYSTEM

Fuel	Plant Site	Boiler Capacity	Type of Catalyst	N/R	Time of Delivery
LNG	Kyushu Electric, Shinkokura 3	600 MW	Pellet	N	6, 1978
	Kyushu Electric, Shinkokura 4	600 MW	Pellet	N	6, 1979
	Osaka Gas Co., Senpoku Plant	20 t/h	Pellet	N	12, 1976
	Osaka Gas Co., Senpoku Plant	20 t/h	Pellet	N	12, 1976
	Aito Co., Chita Factory	30 t/h	Pellet	N	5, 1978
	Aito Co., Chita Factory	30 t/h	Pellet	N	5, 1978
	Aito Co., Chita Factory	30 t/h	Pellet	N	5, 1978
	Dainippon Ink Co., Sakai Factory	30 t/h	Pellet	N	2, 1980
	TK Plant (LNG & BFG)	395 MW	Grid	R	5, 1981
Oil	Sumitomo Chemical Co., Sodegaura	370 t/h	Pellet	R	1, 1976
	Fuji Oil Co., Sodegaura Refinery	160 t/h	Grid	N	12, 1977
	Tokyo Electric, Yokosuka 4	350 MW	Grid	R	2, 1978
	Kansai Electric, Osaka 1	156 MW	Grid	R	6, 1978
	Chubu Electric, Chita 4	700 MW	Grid	R	11, 1979
	Kansai Electric, Osaka 3	156 MW	Grid	R	7, 1980
	Kansai Electric, Osaka 4	156 MW	Grid	R	12, 1979
	Kansai Electric, Sakaiko 1	250 MW	Grid	R	7, 1980
	Kansai Electric, Sakaiko 6	250 MW	Grid	R	12, 1979
	Chugoku Electric, Iwakuni 2	350 MW	Grid	R	12, 1980
	Chugoku Electric, Iwakuni 3	500 MW	Grid	N	4, 1981
	Chubu Electric, Atsumi 3	700 MW	Grid	N	12, 1980
	Chubu Electric, Atsumi 4	700 MW	Grid	N	2, 1981
	Chubu Electric, Shinnagoya 3	220 MW	Grid	R	7, 1980
	Kansai Electric, Osaka 2	156 MW	Grid	R	7, 1981
	Kansai Electric, Tanagawa 3	156 MW	Grid	R	8, 1981
	Kansai Electric, Tanagawa 4	156 MW	Grid	R	3, 1981
	Kansai Electric, Sakaiko 2	250 MW	Grid	R	9, 1981
	Kansai Electric, Sakaiko 4	250 MW	Grid	R	2, 1981
	Kansai Electric, Sakaiko 7	250 MW	Grid	R	9, 1981
	KK Plant	600 MW	Grid	R	7, 1982
Coal	Chugoku Electric, Shimonoseki 1	175 MW	Grid	R	4, 1980
	Joban Electric, Nakoso 8	700 MW	Grid	N	12, 1982
	Chugoku Electric, Shinube 1	75 MW	Grid	R	8, 1982
	Chugoku Electric, Shinube 2	75 MW	Grid	R	7, 1982
	Chugoku Electric, Shinube 3	156 MW	Grid	R	6, 1982
	KM Plant	156 MW	Grid	R	3, 1983

Remarks: N/R N: New unit
R: Retrofit

TABLE II
DESIGN CONDITIONS—SHIMONOSEKI NO. 1 SCR SYSTEM

Boiler	
Type	Mitsubishi—C-E Controlled Circulation® boiler-CCRR
Evaporation	540 t/h
Fuel	Australian bituminous coal and oil
Unit capacity	175 MW
NO_x Removal System	
Type	Selective Catalytic Reduction
Gas flow	550,000 Nm^3/h
Reactor	Vertical down-flow fixed bed
Catalyst	Grid type—10 mm pitch
S_v nominal	3000 H^{-1}
Inlet NO_x concentration	500 ppm
NO_x removal efficiency	51%
Ammonia slip (target)	5 ppm max.
Dust concentration	about 20 g/Nm^3

TABLE III
DESIGN CONDITION OF AN SCR SYSTEM FOR A 500-MW COAL-FIRED BOILER

	Case I	**Case II**
NO_x value at system inlet	500 ppm	500 ppm
NO_x value at system outlet	100 ppm	50 ppm
NO_x removal efficiency	80%	90%
Ammonia slip	less than 5 ppm	less than 5 ppm
Catalyst life (target)	24 months	24 months
Mole ratio (NH_3/NO_x)	0.81	0.91
Gas conditions leaving the boiler		
Gas flow rate	1,553,000 Nm^3/H	
Gas temperature	380 °C	
Oxygen	4.3%	
Sulfur dioxide	1,350 ppm	
Sulfur trioxide	14 ppm	
Particulate	15 g/Nm^3	

TABLE IV
SCR SYSTEM REACTOR CHAMBER SPECIFICATION FOR A 500-MW COAL-FIRED BOILER

		Case I	**Case II**
Type		Fixed bed	Fixed bed
Number		Two	Two
Direction of gas flow		Vertical down	Vertical down
Size	Width	12.4 m	12.4 m
	Height	8.4 m	11.0 m
	Length	10.2 m	10.2 m
Catalyst volume		404 m^3	480 m^3
Catalyst shape		Grid—7 mm pitch	Grid—7 mm pitch
Number of layers		Three	Four
Superficial gas velocity		5.5 m/s	5.5 m/sec
Draft loss thru reactor		less than 75 mm H_2O	less than 90 mm H_2O